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SELECTED TOPICS:

TRANSURANIUM ELEMENTS IN THE GENERAL ENVIRONMENT

June 1978

U. S. Environmental Protection Agency
Office of Radiation Programs
Criteria and Standards Division
Washington, D.C. 20460

PREFACE

On November 30, 1977, the Environmental Protection Agency announced in the Federal Register proposed Federal radiation protection guidance on dose limits for persons exposed to transuranium elements in the environment. In support of this action, the Office of Radiation Programs also published a summary report "Proposed Guidance on Dose Limits for Persons Exposed to Transuranium Elements in the General Environment" (EPA 540/4-77-016, September 1977).

It is the object of this publication to make generally available selected reports that, because of length or complexity, were not included in the summary report. These reports contain additional material on the transuranium elements with respect to movement through environmental pathways leading to the exposure of individuals, dosimetry models, potential biological effects following inhalation and ingestion, environmental sampling and radiochemical procedures, and the technology and costs for cleaning up contaminated land areas.

Comments on these documents would be appreciated. These should be sent to the Director, Criteria and Standards Division (AW-460), Office of Radiation Programs.

W. D. Rowe, Ph.D.
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for Radiation Programs

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ENVIRONMENTAL CHEMISTRY OF PLUTONIUM

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February 1978

U.S. Environmental Protection Agency
Office of Radiation Programs
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Washington, D.C. 20460

1. General Properties

Plutonium is a silvery white metal which readily oxidizes in air. The oxidation rate of plutonium depends upon the temperature and the relative humidity of the air and increases as the relative humidity increases. In going from 0% to 50% relative humidity, the oxidation rate will increase by several orders of magnitude.

Plutonium is capable of forming many compounds with oxygen, including the simple binary oxides, the peroxide, the hydroxide (or hydrated oxides), and a series of ternary and quarternary oxides. Plutonium dioxide (PuO_2) is the most stable of the oxides and is formed under most conditions, especially when plutonium metal is ignited in air. PuO_2 when calcined is highly unreactive under normal environmental conditions, is extremely insoluble in water, and if ingested would be highly insoluble in body fluids. The dioxide is the most common form of plutonium encountered commercially. Because of its desirable properties such as high melting point (2240 °C), irradiation stability, compatibility with metals, chemical stability ($\Delta F_{298} = -253$ kcal), and low vapor pressure, plutonium dioxide has found widespread use as a fuel either alone or in combination with other compounds. For example, plutonium-238 is used as a heat source ($^{238}PuO_2$) for thermoelectric generators employed in devices such as heart pacemakers and communication satellites. When used as a heat source, the $^{238}PuO_2$ is fashioned into pellet form and then encapsulated in a refractory metal. Other solid forms of plutonium are possible but are of little concern from the standpoint of being a likely environmental contaminent.

2. Chemical Behavior in Aqueous Environments

Plutonium dioxide is a highly refractory material when prepared at high temperatures and has been found to react slowly with aqueous solutions. The rate at which dissolution procedes depends on such factors as the pH and temperature of the solution and the presence of oxidizing, reducing, and complexing agents. As the dioxide dissolves, plutonium ions are released which can undergo complexation and/or hydrolysis with other species present in the aqueous phase. Actually hydrolysis can be looked upon as a form of complexation in which the hydroxyl ion is the ligand. The most stable hydrolysis product of plutonium in the pH range generally encountered in natural waters is $Pu(OH)_4$ (Andelman, 1970). This hydroxide has been reported (Taube, 1964) to have a solubility product constant of 7×10^{-56} which is indicative of a high degree of insolubility.

Potentially four oxidation states of plutonium (III, IV, V, VI) can exist in solution in equilibrium with $Pu(OH)_4$. The predominant soluble form of plutonium in a soil/water environment will be determined by the oxidizing or reducing capability of the solution. The availability of plutonium for uptake by plants and other biological systems will vary depending upon the valence state (Adams, 1975 and Price, 1973). Generally, the availability to plants follows the valence order V > VI = III > IV.

Most efforts to ascertain the valence of soluble plutonium have been theoretical because of the difficulty encountered in determining valence states at typically low environmental concentrations. One means of predicting the predominant plutonium species under specified conditions is the use of stability diagrams (Polzer, 1971). These diagrams examine the phase relationships between possible plutonium species as a function of the redox potential of the solution. Their use is limited, however, to systems which are in equilibrium and for which the concentration of additional ions is insignificant. Also, this approach only considers the thermodynamics of the system and not the kinetics, thereby indicating which reactions are posssible but not how fast they will occur. Within these limitations, stability diagrams can be useful in evaluating the observed environmental behavior of plutonium.

From stability diagrams, Polzer (1971) and Andelman (1970) both concluded that, when the concentration of complexing anions is insignificant, the predominant form of soluble plutonium will be Pu^{+3}. However, natural environments contain complex materials from the decomposition of plant and animal matter which may affect the chemical equilibrium of the soil/water environment. Bondietti (1975) states that the presence of a complexing agent such as EDTA can increase the susceptability of Pu^{+3} to oxidation to Pu^{+4} due to the activity-lowering effect that complexation produces. His studies have attempted to ascertain the probable valence state of plutonium in the presence of various organic substances commonly encountered in the environment. He concluded that the Pu(IV) valence state would be the most important and that higher oxidation states would be reduced to the IV state through the action of phenolic materials like humic substances and reducing sugars like uronides. Further valence reduction to Pu(III), although possible, was not felt to be likely under the redox conditions normally encountered. Bondietti observed that only a small fraction (< 20%) of the plutonium in a soil contaminated 30 years previously was desorbable and, therefore, available for uptake by biological systems. The majority of the plutonium was strongly associated with the solid phase and, as a result, leaching losses were concluded to be insignificant compared to the physical movement of contaminated soil particles.

2.1 Freshwater Environment

The above observation is consistent with studies of plutonium released into aquatic systems. For example, Wahlgren (1973) has shown that 95% of the plutonium added to Lake Michigan as a result of atmospheric fallout from weapons testing is rapidly removed from the water column to the sediments. Further studies of Lake Michigan by Alberts (1974) have demonstrated that plutonium is strongly associated with the sediments and is not easily solubilized under aerobic conditions. Stagnant, polluted systems however could have a greater solubilizing effect due to high concentrations of complexing organic material.

Studies of the fate of plutonium in flowing water systems have been conducted in the Great Miami River (Ohio) where discharges of plutonium from Mound Laboratory have occurred. These studies (Bartelt, 1975 and Muller, 1977) have attempted to elucidate the mobility of plutonium in aquatic ecosystems. Consistent with the Lake Michigan studies, plutonium was found associated with the sediments and that at least 90% of the radioactivity which moves down the river does so as a result of the resuspension/entrainment of bottom sediments.

The predominant chemical form of soluble plutonium is, however, still open to question. For example, Alberts' Lake Michigan study indicates that most of the soluble fraction is in an anionic form with a particle diameter of less than 30 Å . This may indicate sorption of plutonium to colloidal silica and other negatively charged colloidal minerals. There has also been speculation that the VI valence state can be an important environmental species. The VI valence state can be generated by the disproportionation of PuO_2 during dissolution (Cleveland, 1970):

$$3\ Pu^{4+} + 2\ H_2O \longrightarrow 2\ Pu^{3+} + PuO_2^{2+} + 4\ H^{+}$$

Although simple stability diagrams do not predict the VI valence to be important, the presence in natural waters of complexing agents (e.g. carbonates) can stabilize this valence state (Andelman, 1970). One such reaction which might associate plutonium in anionic form is:

$$PuO_2^{2+} + 2\ CO_3^{2-} \longrightarrow PuO_2(CO_3)_2^{2-}$$

Even though the chemical behavior of plutonium in fresh water is not completely understood, the many studies to date have consistently demonstrated that plutonium will quickly associate with the solid phase and, under the redox conditions normally encountered in the environment, solubilization will be minimal. As long as the water body is maintained at a fairly constant level and large areas of the sediment bed are not exposed and allowed to dry, thereby becoming a wind resuspension problem, the plutonium activity committed to the water will be bound there in a form which is relatively insoluble in biological systems.

2.2 Marine Environment

Because of its long radiological half-life, it must be assumed that some of the plutonium in any freshwater body will eventually be carried to an ocean or sea. Knowledge of the behavior of this radionuclide in a marine environment is therefore important when evaluating the long term impact of any release to the environment.

In a study conducted off the coast of Massachusetts by Bowen et al., the bioturbation and sedimentation rate effects on plutonium sediment profiles were examined (Bowen, 1975). Analyses of cores taken prior to 1964 and from 1968 to the present showed a progressive migration of Pu from deeper sediment layers to areas nearer the surface. It appears that primary amines emanate from the deeper anoxic or nearly anoxic sediment layers. These amines seem to be involved in a mechanism for releasing Pu from the sediments, but when they reach the oxygenated upper sediment layers what appears to be microbiological consumption immobilizes the Pu again. In this study the remobilization of Pu is attributed to a reducing mechanism, as it was in other studies (Edgington, 1975 and Alberts, 1975). This would indicate a redistribution of Pu in the sediments without a release to the water column. The distribution of Pu in sediment samples with respect to particle size was noted as being uniform. No preference for the finer fractions was observed and concentrations of these nuclides per gram of sediment varied only by a factor of two from the finest to the coarsest fractions measured.

The activities of bottom feeders can also have an effect on sediment Pu profiles. These fish tend to enhance the mixing of the sediments and the water column. This will either increase the deposition rate of the Pu from the water column to the sediments or, in times of relatively low fallout rates, it can enhance possible resolubilization of the sediment bound Pu. However, extremely low and variable Pu levels in the water itself seem to indicate that the Pu inventory is mostly removed to the sediment bed.

In another study, the fate of plutonium released to the Irish sea over approximately 20 years has been examined. This study

(Hetherington, 1975) has noted that > 96% of the plutonium released was removed to the sediments in a relatively short time period. The mechanism responsible for carrying the Pu to the sediment bed does not appear to be biological nor does it seem to be a simple diffusion mechanism. Sorption to particles in the water column and subsequent deposition in the sediment bed appears to be the primary mechanism for removing Pu from the water. Once these sediments are consolidated, there appears to be no detectable remobilization of the plutonium.

Similarly, a study of Bombay Harbor Bay (Pillai, 1975) observed that 99% of the released Pu was localized in the sediments around the discharge point, but areas of higher activity were found in locations with particularly high siltation rates. The Pu-containing sediments were examined to determine the conditions necessary to release the Pu from the sediment. Violent agitation for 8 hours did not effect any detectable release of Pu to seawater. Also, extraction tests using hydrochloric acid at 80 °C resulted in no release of Pu to the solution.

As with freshwater systems, the marine studies have shown that over 90% of ~~the~~ Pu entering either as fallout or in the form of liquid effluents - was found trapped in the sediments. This behavior is mostly attributed to the bonding of Pu to the organic material associated with sediment particles. A very small fraction of the Pu entering a body of water remains unfilterable. Most of this is assumed to be Pu in some colloidal form.

Even though Pu is strongly bound to the sediment bed in most water bodies it can still move with water currents and other physical actions. The effects of biota on Pu behavior are seen mostly in the sediments. Both microorganisms and more complex species can alter the depth profiles. Primary amines released deep in the sediment by microbes can solubilize the Pu in anaerobic regions and carry it towards the surface. Microbes in the anaerobic interface regions, along with chemical reactions, bind the Pu to the sediments again. Larger biota can physically alter profiles by digging into the bed and allowing the overlying water to penetrate. The most significant role of biota is the concentration of Pu in their systems. The most notable cases are seaweeds and shellfish. The seaweeds, with concentration factors of up to 1000 are by far the most notable. The importance of shellfish is their possibility of being a more direct food source, even though they have a lower concentration factor.

3. Chemical Behavior in Soils

Similar to its aqueous behavior, the interaction of plutonium with soils is not completely understood and has been developed to date only qualitatively. This is in part due to the difficulty in characterizing plutonium at very low environmental concentrations. Oftentimes the predominant valence state of plutonium cannot be established-much less its chemical form. However, several studies conducted at sites where plutonium has been in the environment for as long as 30 years have yielded some knowledge of its behavior. One such series has been conducted by Tamura at four sites having different ecosystems. The selected sites were: the Nevada Test Site (Nevada), Oak Ridge National Laboratory (Tennessee), Mound Laboratory (Ohio), and Rocky Flats (Colorado). The source terms were different at the four sites: at NTS the source was a "safety shot" series which dispersed plutonium on the oxide, at ORNL an earthen dike gave way releasing material from a holdup pond, at Mound Laboratory an acid solution of plutonium leaked from a waste transfer line, and at Rocky Flats cutting oil contaminated with plutonium leaked from storage drums. The association of plutonium with the soils of these various sites will be affected by factors associated with the source term and the prevailing local environmental conditions.

The ease in which plutonium is extracted from the solid phase can indicate its availability for biological uptake. Studies by Tamura (1976) have shown low extraction (10-15%) from soils of the Nevada Test Site and Rocky Flats, while ORNL and Mound Laboratory soils showed much higher extraction (60-85%). These results are evidence of the existence of different chemical species at the various sites. Experiments by other investigators have provided further evidence of such differences. Specifically, autoradiographic analyses of soil samples from Rocky Flats (Hayden 1974; Nathans 1974; Sehmel 1975) have shown the presence of discrete particles of plutonium (probably the oxide) attached to larger soil particles, while the same technique indicated a general dispersion of plutonium in the soil of Mound Laboratory (Rodgers 1975).

In order for plutonium to be incorporated into the soil matrix, it must be available in ionic form and capable of displacing some other cation from the matrix. This could explain the difference in the extraction and autoradiography studies at the various sites. At NTS and Rocky Flats the plutonium was originally released as metal or oxide particles which require aqueous dissolution to generate plutonium ions. Contrastingly, the Mound release was as an acid solution already containing plutonium in ionic form which probably reacted quickly with the solid phase and was incorporated into the soil matrix. The Rocky Flats and NTS soils, on the other hand, could contain hydrated or polymerized plutonium oxide attached to soil particles via adhesion. Once polymerized, plutonium is slowly depolymerized by strong acids (Cleveland, 1970) and this could account for the low extractability of the Rocky Flats and NTS samples.

Rodgers (1975) in his studies around Mound Laboratory proposed a mechanism for the bonding of plutonium to soils and the following has been excerpted from his review:

> Proposed bonding mechanisms: Basically, soils are made up of silicate materials and other minerals. Quartz sand has a continuous silicate structure where each silicon atom is bound to two silicons. This continuous structure is interrupted at the surface and in natural systems (where water is abundant), the surface is composed of unsaturated oxygen bonds. That is, each surface oxygen is bound to only one silicon atom. The remaining bond is usually occupied by other cationic species

Clays are more complicated, but are also based on the continuous silicate structure except that some silicon atoms have been replaced by Mg^{2+}, Al^{3+}, Fe^{3+}, and Fe^{2+} (mostly Al^{3+}). These substituted atoms in the silicate structure result in variations in long range crystal structure. Rather than forming three dimensional silicate networks as in quartz, many clays form two-dimentional sheets which cleave easily to form plates. The surface of these layered sheets that make up the clay particles also exhibit unsaturated oxygen bonds. This, in part, accounts for the higher sorption capacity in clays (relative to the same size silicate particle) since sorption can take place between the silicate layers within the clay particles.

The unsaturated oxygen bonds in natural soils and clays are occupied by cations such as H^+, K^+, Ca^{2+}, Mg^{+2}, or other available cations. The bonding strength order of these cations is:

$$Pu^{4+} > H^+ > Al^{3+} > Ba^{2+} > Ca^{2+} > Mg^{2+} > NH_4^+$$
$$K^+ > Na^+.$$

In order to bond to the silicate, an ion must displace or exchange with the cation already bonded:

$$A\text{-Clay} + B^+ \longrightarrow B\text{-Clay} + A^+.$$

The extent of the exchange depends on the relative strength of the bonds and the relative solution concentrations of the two cations.

Some cations form silicate bonds that are fairly weak (such as Na^+ and K^+) and may be only electrostatic while other metal cations may even develop covalent character.

Tetravalent plutonium ions are well noted for the formation of strong bonding (complexing) with oxygenated ligands. The strength of plutonium oxygen bonds is also indicated by the acidic character of plutonium hydroxide forming hydrous plutonium oxide.

It is not surprising then that plutonium ions can compete with hydrogen ions for the bonding sites on the silicates even when the Pu^{4+}/H^+ concentration ratio is 10^{-11} or less. The very large bonding potential of plutonium suggests that sorbed plutonium cannot be significantly displaced from soil by the concentrations of cations existing in nature.

Chemicals that complex the plutonium compete with the silicate particles for the plutonium and tend to reduce the extent of sorption of the plutonium on soil. For example, the formation of plutonium hydrolytic species $PuOH^{3+}$, $Pu(OH)_2^{2+}$, $Pu(OH)_3^+$, and $Pu(OH)_4$ (as well as "polymeric" forms tends to reduce ion exchange sorption. Some of the hydrolytic plutonium "polymeric" forms may adsorb to the surface of the soil particles and the precipitation of $PuO_2 \bullet (H_2O)$ may be nucleated by the colloidal soil particles when the plutonium concentrations are relatively high ($> 10^{-6}$ M). Also moderately strong organic complexing agents, such as citric acid, can reduce the sorption of plutonium on soil.

4. Interaction with Plants

As discussed previously, a consideration (Bondietti, 1975) of the oxidation-reduction potentials of the valence state couples in the presence of organic material ubiquitous to soil indicates that Pu(IV) is the predominate valence state in the environment. Since the availability to plants generally follows the order VI $>$ VI $=$ III $>$ IV, a low uptake of plutonium by plants would be expected. This expectation has been confirmed by the numerous studies conducted in the laboratory and in the environment where many varieties of plants have been grown in contaminated soils. The interested reader is referred to several recent reviews (Bernhardt, 1976; Bulman, 1976;

Mullen, 1976; and Thomas, 1976) which describe such details as the types of crops grown, the amount and chemical form of the plutonium used, and the method by which plutonium was added to the growing medium.

The results of these studies are usually expressed in terms of a "concentration factor" defined as the quotient of the plutonium disintegrations per minute per gram of plant material and plutonium disintegrations per minute per gram of growing medium, with the dry weights of the plant and growing medium usually used. This index indicates the degree to which plutonium is concentrated or discriminated against in the soil to plant pathway. Even though a variety of methods and conditions have been used in these studies, the concentration factors are generally of the order of 10^{-4} to 10^{-5} On occasion some higher values, ranging as high as 10^{-1}, have been reported but these are usually attributed to external contamination of the plant surface rather than incorporation into the plant tissue.

Such small concentration factors tend to support the conclusion that the terrestrial food chain does not constitute a significant exposure pathway. However, the observation has been made that these experiments were all of a short-term nature and, as such, did not consider the possibility of increases in concentration factors over time due to a variety of possible influencing factors. As a result preliminary investigations of several mechanisms of increased uptake have been reported (Cleveland, 1976; Bondietti, 1975; Romney, 1970; Lipton, 1976; Price, 1973; and Beckert, 1975). Included among these are: 1) chelation by organic constituents in the soil or through the addition of fertilizers, 2) increased root absorption zone contact as plutonium migrates down through the soil, 3) increased radiocolloid size due to aging, and 4) the long-term action of microorganisms present in the soil.

Much of the research into these mechanisms is in the embryonic stage, however, the preliminary data have not demonstrated that a substantial transformation will occur under conditions generally encountered in the environment. Most likely, if these transformations take place, they will occur slowly over the order of years to decades. One possible exception might be the chlorination of drinking water. A study investigating this possibility demonstrated a rapid oxidation of Pu(IV) to the more soluble Pu(VI) as a result of chlorination (Larsen, 1976).

5. Conclusions

Although many questions concerning the environmental behavior of plutonium are still unanswered, some conclusions can be drawn from the information currently available. To date all indications are that when plutonium crosses a biological membran e it is discriminated against (with the exception of some marine plants) and that bioaccumulation does not occur though the food chain. Most assessments of the doses from environmental plutonium have concluded that the principal exposure pathway is via the suspension of contaminated soil particles with subsequent inhalation of these particles. The inhalation pathway is then followed in importance to a much lesser degree by the ingestion of water and foodstuffs. However, because of its long radiological half-life and persistence in the environment, such assessments should take into consideration possible long-term changes in the solubility characteristics of plutonium, as well as the ingrowth and mobility of its radiological decay products before discounting the importance of the ingestion pathway.

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A REVIEW OF MODELS FOR THE RESUSPENSION OF PLUTONIUM INTO AIR FROM SOIL SURFACES

George V. Oksza-Chocimowski

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Office of Radiation Programs - Las Vegas Facility
Las Vegas, Nevada 89114

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PREFACE

The Office of Radiation Programs of the U.S. Environmental Protection Agency carries out a national program designed to evaluate population exposure to ionizing and non-ionizing radiation, and to protect the public health and safety by promoting development of the necessary controls.

The present report comprises a review and evaluation of presently available analytical, empirical, and computerized models intended to predict the redistribution of plutonium following initial ground deposition, with special emphasis being placed on resuspension.

Readers of this report are encouraged to inform the Office of Radiation Programs of any omissions or errors. Comments or requests for further information are also invited.

Donald W. Hendricks

Donald W. Hendricks
Director, Office of Radiation
Programs, Las Vegas Facility

ABSTRACT

Presently available models of plutonium resuspension are presented and discussed in this report. The models include 1) time-dependent resuspension factors (Langham, 1969; Kathren 1968; Anspaugh et al., 1974), 2) Slinn's wind/erodibility-dependent factor (1975), 3) mass-loading, 4) Sehmel and Orgill (1973) wind-dependent airborne concentration, 5) friction velocity-dependent dust flux/concentration, (Gillette and Shinn, 1974), 6) atmospheric diffusion [Healy, Fuquay (1959, 1974)], 7) Horst et al. analytical model (1974), 8) computerized approaches [Amato, Travis (1975)]. The relationships of some of these models are investigated and discussed, as well as various factors affecting resuspension.

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INTRODUCTION

For historical reasons, the concern with contaminants discharged to the atmosphere has been traditionally limited to the material during the course of dilution, before deposition had occurred. However, with nuclear power quickly becoming a valid alternative to rapidly depleting traditional sources, other aspects of the contamination problem have to be considered.

Effluents from operating nuclear facilities are likely to be radioactive, to an extent varying with the nature of the release (expected or accidental) and the type of facility. A "cloud" of such effluents, affected by atmospheric motions in the same manner as would be more common pollutants, would nevertheless pose additional hazards, not encountered in the latter case.

Conceptually, two elements of risk are involved. The first element is the danger of direct radiation exposure and inhalation of radioactive substances during cloud passage. The second element is the radiation insult on a long-term basis from the radioactive material deposited during cloud passage. This second element includes direct radiation exposure, inhalation of resuspended material, and ingestion of food contaminated by the deposited material. Under nominal conditions, and depending on the isotopic inventory of the effluent, the exposure to γ-radiation and breathing hazard associated with long-term occupancy of an area contaminated by deposition from the passing cloud may be comparable to that resultant from the cloud passage.

After deposition of the radioactive particles on ground surfaces, vegetation, or other obstructions in their path, these particles may be moved or again become airborne, "resuspended", through the action of wind forces. When the contamination is due to long-lived radioactive materials which do not give rise to a significant external radiation field, the inhalation hazard due to resuspension is particularly important. Such materials are those containing alpha active species and include plutonium, uranium, and polonium.

Of particular interest is plutonium-239, because of its unique usefulness as a reactor fuel in breeder reactor concepts. Plutonium-239 (half-life 24,400 yrs) normally decays by emission of alpha particles of an average energy of 5.15 MeV, with additional soft X-rays (17-20 and 60 KeV) being given off either by impurities or decay products. One microgram of pure plutonium-239 gives off approximately 140,000 alphas per minute, which represents a specific activity of 60 mCi/g (Langham, (1969a).

Upon release, either expected or accidental, from a nuclear facility, plutonium would be dispersed in the form of fine particulates, often as PuO_2, (Fish et al., 1972; Mishima, 1964) which would serve to reduce the specific activity. In addition, the dispersion of relatively large quantities of plutonium oxide over areas large enough to constitute a potential hazard to the local ecosystem would result in much lesser specific activities per gram of contaminated soil. The highest values reported by E. M. Romney, H. M. Mork, and K. H. Larson in a 1970 study, almost eleven years after a high explosive detonation, were equivalent to about 6×10^{-6} mCi/g in the top three centimeters of topsoil. Simultaneously, alpha activity in windblown material indicated that wind erosion had caused considerable redistribution of the original fallout contaminant (Romney et al., 1970). Similar conclusions were reached by S. E. Poet and E. A. Martell in a study of plutonium-239 contamination in offsite areas near the Rocky Flats plutonium plant. Measurements in the top one centimeter surface layer of soils showed concentrations up to hundreds of times that of fallout. Most of this offsite accumulation of plutonium-239 corresponded to a period in which plutonium on the plant site was exposed and subject to wind reentrainment (Poet and Martell, 1972).

These are but two concrete examples of plutonium redistribution as affected by meteorological factors. Conceptually they involve nothing new, inasmuch as any material deposited on or forming part of the upper layer of topsoil is prone to be redistributed by the action of wind forces. Roughly one-tenth of the global mass of atmospheric aerosols is presumed to consist of dust, based on estimates of worldwide wind erosion of soil amounting to $2 \times 10^4 - 10^6$ tons per day (Travis, 1975). A single dust storm on May 12, 1934, removed an estimated 200 to 300 million tons of material, mostly fertile topsoil, from the Great Plains cultivation areas (Hilst & Nickola, 1959). Similar occurrences have plagued agriculturalists the world over since time immemorial.

Some concepts in the field of soil erosion were contributed by Bagnold, Chepil, Hilst, Nickola and many others. Bagnold (1941) described the initiation of particle movement as occurring by either or both of two means:

1. Air moving past a particle resting on a surface may exert sufficient drag to dislodge it from this surface and subsequently cause it to roll, slide or even become airborne by bouncing off other loose particles or minute irregularities of the terrain. Assuming that the particle is sufficiently large for forces of cohesion or adhesion to be ignored (more will be said about them later), it can easily be shown that drag forces may overcome the forces of gravity on a particle resting on the ground.

Drag forces are proportional to a particle cross-sectional area,

$F_D \propto D^2$ where F_D = drag force (1)

D = diameter of particle

Gravity forces are proportional to a particle mass and thus to the particle volume

$F_g = mg = \rho Vg \propto D^3$ where F_g = gravity force (2)

m = mass

g = acceleration of gravity

ρ = density

V = volume.

The ratio of drag to gravity forces is thus inversely proportional to the particle diameter.

$$\frac{F_D}{F_g} \propto \frac{D^2}{D^3} \propto \frac{1}{D} \qquad (3)$$

Consequently, the smaller the diameter of a particle (down to certain limits) the greater the propensity for being dislodged by wind forces.

2. Once airborne, the particle may acquire additional kinetic energy from the air stream and, returning to the surface by the action of gravity, transfer part of the momentum to a stationary particle, dislodging it in

turn. This is the second mechanism of initiation of motion. The falling particle may also do one of several other things:

a. Collide elastically (almost), returning to the air stream

b. Roll or slide along the surface

c. Be absorbed into the surface without further motion

d. Shatter another particle

e. Shatter itself (Hilst & Nickola, 1959)

All of these mechanisms, singly or in combination, involve energy transfer to the soil, a particle, a number of particles and/or a number of particle fragments. Assuming that the rate of particle "pick up" by direct wind forces is maintained, the dislodgment of additional particles by impingement of those previously airborne would result in a rapid increase in the number of moving particles as a function of distance downwind from the site of the initial movement. It has often been observed that erosion of a surface increases downwind from its upwind edge (Bagnold, 1941; Hilst & Nickola, 1959).

It should be pointed out that an airborne particle does not necessarily have to return to the surface shortly after wind pickup. Once airborne, a particle is subject to two forces: the force of gravity, downwards, a function of mass, and the force of air resistance or drag, function of the particle size and velocity, acting in the direction opposite to that of particle motion relative to the surroundings (air stream, in this case). Drag forces on particles of small size may exceed the force of gravity by several orders of magnitude, thus forcing the particle to conform very closely to the motion of the air stream in which it is carried.

Drag force in air $F_D = C \times \frac{1}{2} \rho a v^2$ (4)

where C = constant, experimentally determined

ρ = density of particle

a = cross-sectional area of particle

$= \frac{\pi D^2}{4}$ where D = diameter.

v = relative velocity of particle in air stream

Recalling that gravitational force $F_g = mg = \rho V g$ where V = volume = $\frac{1}{6} \pi D^3$, a ratio defining the propensity of a particle to remain suspended may be established:

$$\text{"Susceptibility"} = \frac{\text{Drag force}}{\text{Gravitational force}} = \frac{C \times \frac{1}{2} \rho \frac{\pi D^2}{4} v^2}{\rho \frac{1}{6} \pi D^3 g} \quad (5)$$

$$= \frac{3}{4} \frac{C}{D} \frac{v^2}{g} \quad (6)$$

Again, the smaller the particle diameter the greater the "susceptibility" and the particle tendency to remain aloft. Thus the ratio of these two forces in air is not altogether different from their ratio with the particle resting on the ground. In the latter case the relative velocity was assumed to be that of the air stream,constant for a given set of conditions. In air, relative velocity is subject to change, being initially close to the air stream velocity but decreasing in time to a value known as "terminal velocity," corresponding to unit susceptibility. This is the velocity with which smaller particles will return to the ground, unless impeded by updrafts effecting further changes in their relative velocity. Thus, a small particle may travel many miles before touching ground. Larger particles, under the same conditions, may return to the surface shortly after "pickup," without ever achieving terminal velocities, by virtue of their small susceptibilities. Consequently, the size of the particles will determine, within some rather nebulous limits, their general mode of transport, defined in Table 1 (Bagnold, 1941; Hilst & Nickola, 1959).

TABLE 1. MODE OF TRANSPORT AS FUNCTION OF PARTICLE SIZE

Mode of Transport	Description of Motion	Particle Diameter
Surface creep	Sliding or rolling along	"large," D >1000 μm
Saltation	By leaps and bounds, in short hops along surface, at a small distance from the ground surface.	"intermediate," 50 μm <D <1000 μm
Suspension	Airborne motion following air stream.	"small," D <50 μm

The relative proportions by mass of the three modes of soil transport vary widely on different soils. Surface creep has accounted for 7 percent to 25 percent of total flow, depending on soil type. Saltation flow varied accordingly, between 55 percent and 72 percent, most of it below the height of 2 or 3 feet. Flow in suspension showed a somewhat greater range of values, from 3 percent to as much as 38 percent, in rough correspondence to the proportion of particles smaller than 100 micrometers in the soil (Chepil, 1945a). The source gives tables of average results indicating this relationship, summarized below by means of a table and graph.

TABLE 2. SOIL FRACTIONS RESUSPENDED AND AVAILABLE FOR RESUSPENSION *

Soil Type	Percent of soil Removed in Suspension	Percent of Particles <100 µm in the soil
Sceptic Heavy Clay	3.2	1.5
Fine Dune Sand	16.6	8.0
Hatton Fine Sandy Loam	32.2	26.0
Haverhill Loam	38.1	32.0

* After W.S. Chepil, "Dynamics of Wind Erosion: I Nature of Movement of Soil by Wind," Soil Science, Vol-60 (1945)

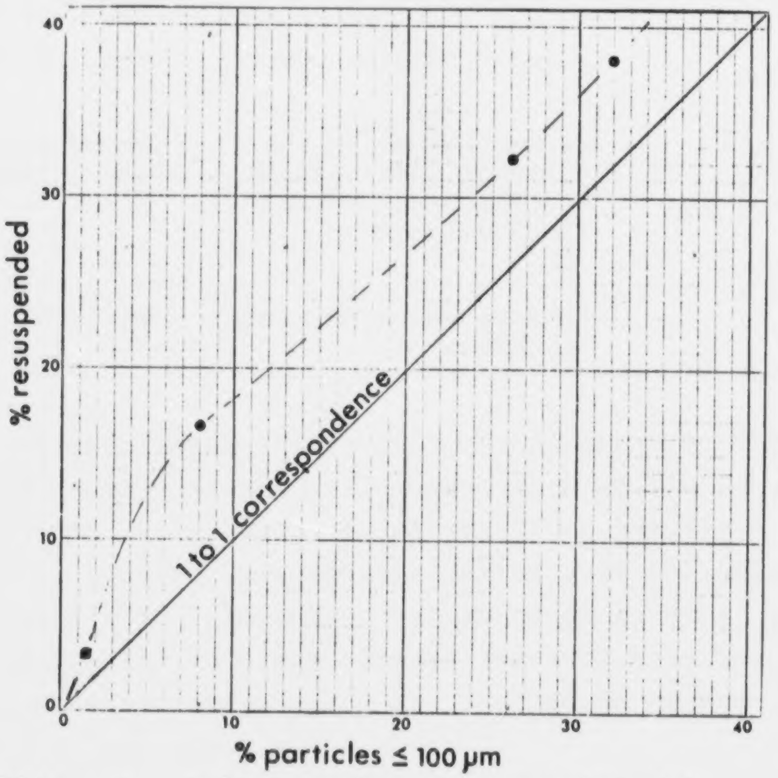

Figure 1. Soil Fractions Resuspended and Available for Resuspension

Obviously some particles larger than 100 micrometers were carried in resuspension. To obtain a closer correspondence of particles percent in the soil to suspended fraction, the limits on particle sizes susceptible to resuspension may have to be revised. The great number of intervening factors affecting soil erosion (and thus resuspension) should suggest how speculative these limits are. Some of these factors have been listed by G. R. Hilst and Nickola and some by W. S. Chepil, (see Table 3).

Presumably, an accurate prediction of the amount of erosion at a certain location at a given time would involve consideration of all the above factors. This would mean, naturally, that the effect of any one of these factors, suitably parameterized, and in combination with average wind speed, would have to be determined independently of any other, i.e., holding all others constant. The difficulties of physically achieving this are obvious.

There exist numerous empirical relationships of sometimes uncertain limits of applicability, each relating several of these factors to soil transport. However, by the very nature of the process followed in their derivation, the effect of other variables had to be neglected or included by means of "constant" factors varying discretely with gross changes in the variables not of interest. That these factors would have to be applied in order to maintain the applicability of the equation over certain range of conditions would tend to obscure the true importance of the variables under study. The magnitude of the effects presumably due to one defined parameter may thus completely overshadow the influence of others, conceivably of great importance under different conditions. Even parameterizing some of these factors can be a project of great complexity, plagued by a multitude of different approaches with differing concepts and methods of measurement. In addition, the paucity of reliable physical equations makes successful dimensional analyses improbable.

TABLE 3. FACTORS INFLUENCING WIND EROSION

AIR (1)	GROUND (1)	SOIL (1)
Velocity	Roughness	Structure affected by:
Turbulence	Cover	Organic Matter
Density affected by:	Obstructions	Lime Content
Temperature	Temperature	Texture
Pressure	Topographic Features	Specific Gravity
Humidity		Moisture
Viscosity		

SURFACE PROPERTIES (2)

- Large-scale surface roughness
  - Mechanical turbulence
  - Overall sheltering
- Small-scale surface roughness
  - Sheltering of individual particles
- Area of erodible surface
- Vegetative cover
  - Live vegetation
  - Plant residue
- Cohesiveness of individual particles
  - Moisture of surface
  - Binding acting of organic materials

PARTICLE PROPERTIES (2)

- Particle size frequency distribution
  - Ratio of erodible to nonerodible fractions
- Particle density
- Particle shape

METEOROLOGY FACTORS (2)

- Wind velocity distribution in the surface layer
  - Mean wind speed
  - Wind direction
  - Frequency, period, and intensity of gusts
  - Vertical turbulent exchange

- Moisture content on ground surface
  - Precipitation
  - Dew and frost
  - Drying action of the air

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(1) W. S. Chepil, "Dynamics of Wind Erosion: I Nature of Movement of Soil by Wind," 1945.

(2) G. R. Hilst and P. W. Nickola, "On the Wind Erosion of Small Particles," 1959.

Consequently, attempts at modeling erosion processes, in particular resuspension, have been limited to relating several grossly measurable variables by means of factors or strictly empirical expressions which may reflect no more than a circumstantial relationship, thus bypassing the complications inherent to the use of parameters of greater physical significance. Paradoxically, the very lack of physical meaning of the parameters used may eventually lead to their applicability to a wide range of conditions, such as often found in gaging resuspension over wide areas, in which the factors listed in Table 3 could show great variability, both spatially and temporally.

RESUSPENSION FACTORS

1. LANGHAM'S MODEL

Evidently, one of the earliest attempts to produce a simple means of predicting the extent of resuspension of pollutant from a contaminated surface dates back to 1956. P. S. Harris and W. H. Langham correlated surface deposition and air concentration measurements of plutonium at the Nevada Test Site (NTS) by use of a quantity known as the "resuspension factor", R_F

$$R_F \cong \frac{\text{Air Concentration } (\mu\text{Ci plutonium/m}^3)}{\text{Surface Deposition } (\mu\text{Ci plutonium/m}^2)} = 7 \times 10^{-5}\ \text{m}^{-1} \quad (7)$$

In addition, an "attenuation factor", λ_A, was calculated to describe the exponential decline in air concentration with time due to progressive reduction of the amount available for resuspension. For NTS conditions, this corresponded to a half-time of 35 days (Langham, 1969a, 1971)

$$\lambda_A \cong \frac{\ln 2}{T(1/2)} = \frac{0.693}{35 \text{ days}} \cong 0.02 \text{ days}^{-1} \quad (8)$$

The processes whereby a pollutant becomes less erodible are commonly grouped under the general term "weathering" (Anspaugh, 1975). They involve the transport of small particles downward into the soil by percolation and cementing of smaller particles into or onto larger ones by the force of adhesion or cohesion.

From the definition of resuspension factor, the air concentration at any time t is

$$C\ (t) = R_F\ S_a\ (t) \quad (9)$$

Where $C(t)$ = air concentration at time t, $\mu Ci/m^3$

R_F = resuspension factor, m^{-1}

$S_a(t)$ = surface concentration at time t, $\mu Ci/m^2$

However, the decrease in air concentration C(t) has been related to contaminant immobilization in the soil by the attenuation factor λ_A.

$$S_a(t) = S_a(o)\ e^{-\lambda_A t} \tag{10}$$

2. KATHREN'S MODEL

A formulation similar to Langham's was used by Kathren (1968), with $T_{1/2}$ = 45 days, commonly referred to as the "Kathren model." In this model, however, the coefficient λ_A is referred to as the "air concentration reduction factor," which implies a different interpretation (Equations (11) and (13) below).

Combining (9) and (10),

$$C(t) = R_F\ S_a(o)\ e^{-\lambda_A t} \tag{11}$$

and since by definition of R_F,

$$C(o) = R_F\ S_a(o) \tag{12}$$

it follows that

$$C(t) = C(o)\ e^{-\lambda_A t} \tag{13}$$

Consequently, the rate of decay of air concentrations may be used to describe the decrease in availability of resuspendible particles in the soil.

3. SURFACE CONTAMINATION AND PENETRATION INTO THE SOIL

It should be pointed out that the term $S_a(t)$ in Langham's and Kathren's models can be misleading, inasmuch as measurements of "surface" contamination are made considering the top layer of soil, i.e., that portion of the total contamination which is available for resuspension. Thus, S_a should be differentiated from total activity existing in the soil per unit surface area S_A, since a fraction of the latter may be due to the radionuclide present at depths greater than that of the easily erodible surface layer. The two quantities, S_a and S_A, can be related once the distribution of contaminant throughout the soil is known.

Beck (1966) and Anspaugh (1975) expressed the distribution of the pollutant in the ground in terms of activity per unit mass of soil, S_m, as an exponential function of depth Z times an empirical constant, characteristic of the soil, known as the "soil profile relaxation constant," λ_p .

$$S_m(Z) = S_m(o)\, e^{-\lambda_p Z} \qquad (14)$$

where $S_m(Z)$ = activity per unit mass of soil at depth Z, $\frac{\mu Ci}{gm}$

$S_m(o)$ = activity per unit mass of soil at surface, μCi/gm

λ_p = relaxation constant, cm^{-1}

Z = depth, cm

The contribution to the total activity per unit surface area S_A of an infinitesimal element of contaminated soil mass extending to a depth dZ is therefore

$$dS_A = S_m(Z)\, \rho\, dZ \qquad (15)$$

where ρ = density of soil, gms/cm^3

Consequently the total activity due to that fraction of the soil extending from the surface to a depth D can be obtained by integration

$$S_{AD} = \int_0^D S_m(Z)\rho dZ \qquad (16)$$

$$= S_m(o)\rho\int_0^D e^{-\lambda_p Z}\, dZ \qquad (17)$$

$$= \frac{\rho}{\lambda_p} S_m(o)(1-e^{-\lambda_p D}) \qquad (18)$$

where S_{AD} = total activity "above D," μCi/cm^2

Obviously, the total activity due to contamination extending from ground level to the farthest extent of penetration of pollutant can be obtained in similar fashion, by integrating from 0 to infinity.

$$S_A = \int_0^\infty S_m(Z)\rho dZ \qquad (19)$$

$$= \frac{\rho}{\lambda_p} S_m(o)\ [1 - 0] \qquad (20)$$

Therefore, that fraction of the activity due to the soil "above D" is (Beck, 1966; Rogowski & Tamura, 1970)

$$Y_{AD} = \frac{S_{AD}}{S_A} = 1 - e^{-\lambda_p D} \qquad (21)$$

where Y_{AD} = fraction of total activity above depth D, dimensionless

D = depth of interest

Choosing D to be the usual depth to which samples of surface concentration S_a are taken, Langham's and Kathren's surface concentration S_a can be related to the total activity per unit area S_A.

$$S_a = S_A Y_{AD} = S_A(1 - e^{-\lambda_p D}) \tag{22}$$

4. SOIL PROFILE AND TIME-INDEPENDENT ATTENUATION FACTOR

The present author judges λ_p to be, obviously, a function of time, since upon initial deposition of the contaminant the entire activity will be concentrated in the top layer of the soil.

At time t = o, $S_a(o) = S_A$ (23)

Consequently, $\lambda_p(o) = \infty$ (24)

As time progresses, the transport of particles into the soil by percolation would establish a certain profile of activity distribution with depth. Thus λ_p is some as yet unspecified function of time, $\lambda_p(t)$, and equation (22) should be properly rewritten as

$$S_a(t) = S_a(o)[1 - e^{-\lambda_p(t)D}] \tag{25}$$

where S_A was replaced with S_a (o) making use of the relationship in equation (23).

Equating the above expression with equation (10), it becomes clear that

$$1 - e^{-\lambda_p(t)D} = e^{-\lambda_A t} \tag{26}$$

and hence $\lambda_p(t) = \dfrac{- \ln (1 - e^{-\lambda_A t})}{D}$ (27)

From the above equation, $\lambda_p(t)$ should be expected to decrease by a factor of 2 with every 35 or 45 days, depending on whether

Table 4. Distribution of residual ^{239}Pu in undisturbed soil profiles

Profile depth (cm)	Profile① (dis/min/g)	Profile② (dis/min/g)	Profile③ (dis/min/g)
0–3	2860 (1.3)*	12,050 (10.8)	6200 (10.8)
3–6	163	2440	1710
6–9	21	344	545
9–12	54	353	215

* Residence time (year) of residual ^{239}Pu when sampled.

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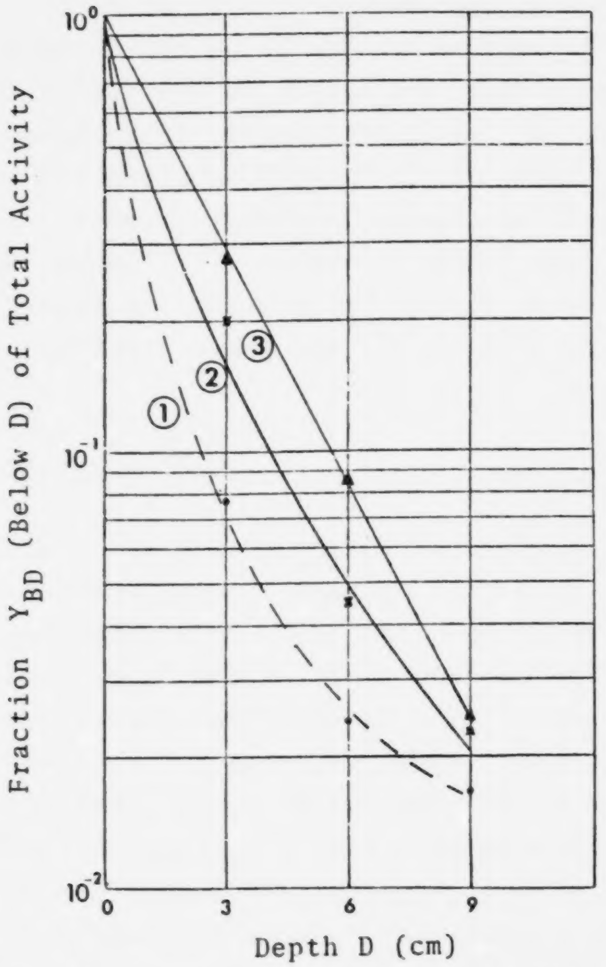

Figure 2. ^{239}Pu Soil Profiles with Time, from Table 4.

Langham's or Kathren's value of λ_A is used. This implies that the pollutant would have to be distributed over twice the depth with every such time interval. As could well be expected, this is hardly the case. Olafson and Larson (1961) noted that plutonium-239 remains very near to the soil surface during long periods of time, and that there was very little evidence of downward migration in the 11 years following the Trinity test in New Mexico. Analyses of profile samples at Yucca Flats showed some downward migration to a maximum depth of two inches, but with little change over a two-year period (Mork, 1970). Vertical profiles obtained by Poet and Martell (1972) at Rocky Flats indicate comparable activities at depths up to 10 centimeters in areas presumably contaminated as result of the 1957 fire, in undisturbed areas east of the plant, and in areas heavily contaminated by the oil spill in 1967, some of which had been ploughed. Data typical of plutonium-239 distribution in profiles of undisturbed soil (see Table 4) obtained by Romney, Mork and Larson (1970) show little difference between distributions after 1.3 years of residence in the soil and those resultant from 10.8 years residence (Figure 2).

5. SOIL PROFILE AND TIME-DEPENDENT ATTENUATION FACTOR

From the preceding discussion in Section 4, it would appear that $\lambda_p(t)$ cannot be related to a time independent attenuation factor λ_A as indicated by equation (26). Nevertheless, that $\lambda_p(t)$ changes with time is undeniable, since from an original magnitude of $\lambda_p(o) = \infty$ at $t = o$ (deposition time) it must decrease to the much lower values measured by the several researchers mentioned below. Furthermore, once these low values are reached little additional change is observed with further passage of time. In effect, Rogowski and Tamura (1970) calculated $\lambda_p = 0.86\ \text{cm}^{-1}$ by fitting data points obtained over a span of two years after deposition to an equation $Y_{BD} = e^{-\lambda_p D}$, where Y_{BD} is the fraction of total activity below a depth of D centimeters (Figure 3).

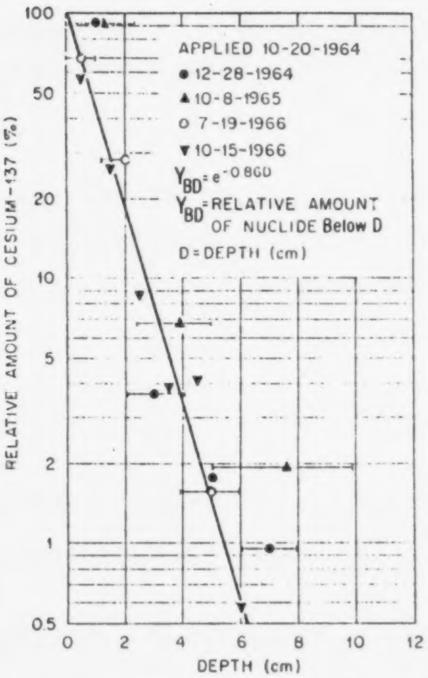

FIG. 3. Relative distribution of ^{137}Cs with depth on Captina silt loam.

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Little difference is apparent between the point distrubution two months after application and that of almost two years later. Thus, not only was a practically constant value of λ_p effective for the two years after deposition, but also, this constant value was very nearly reached in the first two months following the event. Similar conclusions were reached by Beck (1966) on the basis of results obtained by a number of investigators. He decided that "fallout isotopes present in the soil for longer than one month can be reasonably assumed to be distributed exponentially as a function of depth with a relaxation length of approximately 3 cm" ($\lambda = 1/3 \text{ cm}^{-1}$).

What happens in this initial period of one month is crucial in modeling λ_p as a function of time. Summarizing the known facts about the behavior of λ_p, the following points may be restated:

1. At time $t = o$, $\lambda_p(o) = \infty$
2. At time $t > n$ years, λ_p = Constant (a more precise value of n may depend on the soil, amount of rainfall, etc.)
3. At time $t \gtrsim 1$ month, $\lambda_p \equiv$ constant (the degree of approximation to the final constant value depending, again, on local conditions)

Obviously a very rapid drop in λ_p must occur within a short time interval after deposition, to be followed by a much slower rate of change, with λ_p beginning the asymptotic approach to a constant value within a few weeks after initial surface contamination. This suggests, to the author, several possible forms for $\lambda_p(t)$, of which equation (28) is the simplest, though not necessarily the "best," or most accurate.

$$\lambda_p(t) = \frac{A}{t} + B \tag{28}$$

where A,B = constants, controlled by local conditions

t = time

The import of the preceding discussion is that, if $\lambda_p(t)$ is to be related to λ_A, (i.e., if weathering by percolation is to have any significant impact on the attenuation factor) then λ_A should reflect in some degree the time dependent behavior of $\lambda_p(t)$. As pointed out previously, λ_A being constant does not accomplish this goal. Whereas at some given time after deposition Langham's $T_{1/2} = 35$ days or Kathren's $T_{1/2} = 45$ may result in a value of λ_A that corresponds to the appropriate values of λ_p at that time, in general this would not be the case. The need for a time-dependent λ_A would therefore be indicated, possibly as a time function similar to that expressed by equation (28). In fact, if penetration into the soil were considered the principal weathering effect, a tentative expression for $\lambda_A(t)$ could be derived by combining equations (26) and (28):

$$\lambda_A(t) = \frac{-\ln[1 - e^{-(\frac{AD}{t} + BD)}]}{t} \qquad (29)$$

Making use of the series expansion $\ln x = (x-1) - \frac{1}{2}(x-1)^2 + \frac{1}{3}(x-1)^3 - \cdots$, equation (29) may be expressed as:

$$\lambda_A(t) = \frac{e^{-(\frac{A'}{t} + B')} + 1/2\ e^{-2(\frac{A'}{t} + B')} + 1/3\ e^{-3(\frac{A'}{t} + B')}}{t} + \ldots \qquad (30)$$

Discarding the higher order terms in the above equation reduces it to:

$$\lambda_A(t) = \frac{1}{t}\, e^{-(\frac{A'}{t} + B')} = \frac{C}{t}\, e^{\frac{-A'}{t}} \qquad (31)$$

where $C = e^{-B'}$

Applying this expression to the purpose of determining the surface concentration of pollutant available for resuspension produces the following:

$$S_a(t) = S_a(o)\, e^{-C \exp(-A'/t)} \tag{32}$$

Note that for "sufficiently large" values of t the time dependent factor exp (-A'/t) can be simplified by the approximation:

$$e^{\frac{-A'}{t}} \cong (1 - \frac{A'}{t}) \text{ , if } A' \ll t \tag{33}$$

This would reduce equation (32) to a simpler form:

$$S_a(t) \approx S_a(o)\, e^{-C(1 - \frac{A'}{t})} = S_a(o)\, e^{(\frac{A''}{t} - C)} = S_a(o) K\, e^{\frac{A''}{t}} \tag{34}$$

where $A'' = CA'$

$K = e^{-C}$

For time $t \Rightarrow \infty$ (corresponding to an aged source), the relationship becomes:

$$S_a(t) = K\, S_a(o) = \frac{S_a(o)}{e^C} \tag{35}$$

In other words, if soil penetration is assumed to have a significant effect on availability of surface contamination to resuspension processes, the overall behavior of $\lambda_p(t)$ would indicate that $S_a(t)$ should reach a constant value after long presence in the soil surface.

6. EFFECTS OF PARTICLE SIZE ON ADHESION

Another aspect of the "weathering" process whereby a pollutant becomes less erodible involves primarily the soil surface. Following deposition, small particles of resuspendible size may agglomerate into larger units not so easily carried by the air stream, or adhere to bigger particles of a different species or onto portions of the soil that may be considered stationary for all practical purposes. In this respect the behavior of the plutonium contaminant may be no different from that of any other aerosol. There is some evidence that small $^{239}PuO_2$ particles lose their identity as discrete aerosols through attachment to inactive materials in the soil. Even under laboratory conditions minute resuspended particles were found to consist of small amounts of plutonium incorporated into relatively large host particles. Simultaneously with this observation, optical inspection revealed the almost total absence of agglomerates (Moss, Hyatt, Schulte, 1961).

Translating these observations to field conditions, it could well be expected that adhesion to soil particles rather than agglomeration with particles of the same species would characterize the behavior of a deposited plutonium aerosol. This would stem, if for no other reason, from the large numbers of soil particles with which a $^{239}PuO_2$ aerosol may enter into contact when compared to the relatively much smaller concentrations of the pollutant.

In general, adhesion may be considerably enhanced by the process of weathering (now used in the strict geological sense) by which particles are "broken-up" into smaller units that would attach themselves more readily to various surfaces. The strength of this adhesive contact, presumably due to electrostatic, capillary, Van der Waals forces (Walker and Fish, 1967), would increase as the size of the particle decreases (Corn and Stein, 1967). Assuming that the forces of adhesion between differing particles are essentially of the same nature as cohesive forces between grains of the same species, the ratio of these surface forces to gravity would vary inversely with the particle diameter (Terzaghi & Peck, 1968), that is, roughly in the same manner as the ratio of drag forces to gravity forces, discussed previously. This would indicate that drag and adhesive forces could be of the same order of magnitude, at least under certain unspecified conditions, with susceptibility to resuspension then varying according to particle shape, soil conditions, humidity and an imposing array of other factors (some of which are mentioned in Table 3). Another effect of size contributing to the retention of small aerosols on the ground is that they may be too small to protrude above the laminar viscous layer of air close to the surface (Chepil, 1945b).

Thus, somewhat paradoxically, the very size effects that maintain a particle in suspension could make it difficult to resuspend the particle in the first place. Fine silt particles of diameters between 5-10 micrometers did not move even under wind velocities of 37 mph, measured at a 6-inch height. The movement of these small particles, however, would be facilitated when mixed with larger grains capable of moving in saltation, the condition most often found in nature (Chepil, 1945b). Consequently, to preclude resuspension, the particles of interest would have to be quite small, below a certain minimum size, and attached "directly" to the soil surface or to loose particles able to move in creep or saltation but not in suspension. Chepil (1945b) indicated such a minimum size by stating that "particles smaller than [5 micrometers] do not exist as such in ordinary soils, for they are aggregated into larger individual grains." Presumably, the source meant equivalent diameter, in this instance defined as follows (Chepil, 1951b):

$$D_{eq} = \frac{D_{ph}\,\rho}{2.65\ \mathrm{g/cm^3}} \qquad (36)$$

where D_{eq} = equivalent diameter

D_{ph} = physical diameter

ρ = density, grams/cm^3

2.65 g/cm^3 = density of a standard grain of sand

With a density of 11.46 g/cm^3, the physical diameter of a particle of $^{239}PuO_2$ corresponding to an equivalent diameter of five micrometer is approximately one micrometer. Particles of a smaller size to be found in suspension would then have to originate from whatever processes produced the initial pollutant release (explosion, plutonium metal preparation) or from the process of abrasion, whereby grains moving in saltation chip off small particles from indurated materials to which plutonium adhered at some previous time. In the later case the small plutonium-bearing fragments may or may not resuspend, depending on the mechanics of impact and the height of the laminar layer. Chepil (1945c) noted the "negative abrasion" action of moving fine soil particles due to their sticking to various soil surfaces, thus causing an increase in weight of the exposed soil.

Particles of 50 micrometer equivalent diameter, on the other hand, are most easily resuspendible (Chepil, 1945b, 1951a) as evidenced by the extent of erosion of soils consisting of erodible fractions of that size and by studies of the composition of suspended dust (Chepil, 1957a). This would suggest that adhesion forces acting on $^{239}PuO_2$ particles of approximately 10 micrometer in physical diameter are negligible for most purposes. It is hardly necessary to point out that, should these particles disintegrate, adhesive forces would become increasingly important.

7. CONTRIBUTION OF RADIOACTIVITY TO WEATHERING PROCESSES

Extension of the Review, Contributed by the Author

A neglected long-term contribution to the weathering of resuspendible particles of $^{239}PuO_2$ may be provided, in the opinion of the author, by their radioactive nature, to an extent much greater than would be expected from the long half-life of plutonium-239.

Following an alpha emission in a given particle, the remaining 86 keV uranium-235 nucleus is expected to recoil over a range calculated by Fleischer (1975) to be 200 Å (0.02 micrometer). If sufficiently near to the surface of the particle, this alpha-recoil nucleus may escape, ejecting in the process a cone of material assumed to be of radius 50 Å and height 50 Å, containing 10,000 atoms, one-third of which are plutonium-239 atoms. If the cone subsequently becomes attached to a nonsuspendible particle, as is likely, the net result would be that of a reduction of resuspendible material.

The preceding "direct ejection" model is clearly a simplistic one. Alternatively, atomic disarray along the alpha-recoil tracks is the cause of the material removal, possibly due to its enhanced chemical solubility (Fleischer, 1975). The amount of damage along the track was estimated on the basis of alpha-tracks being similar (except for their much shorter length) to those produced by fission fragments. It should be mentioned that fission fragment tracks in LiF have been observed to have widths of about 100 Å and lengths up to 2.3 micrometer (23,000 Å), still less than 25 percent of the total expected range. In UO_2 fuel this total range is approximately 4 micrometer, probably representing the shortest range expected in either the fuel or the LiF crystal (Knorr, 1964). Similar observations made by Noggle and Stiegler (1960) and by Price and Walker (1962) confirm the track widths to be of about 100 Å, prior to chemical etching. These and other fission track data suggested that alpha-track damage would lead to the ejection of volumes corresponding to cylinders of a length of 100 Å (a conservative value), and diameters ranging between 65 Å and 100 Å , resulting ultimately, according to Fleischer, in the loss of 30-80 times as many atoms per event as there would be for the direct ejection model.

In returning to the direct ejection model, an important effect of the particle size on the effective number of atoms ejected per alpha emission needs to be considered. It was noted previously that expulsion of material would follow an alpha-decay event occurring "sufficiently near" to the surface of the particle. The recoil nucleus range (Fleischer's) $D_\alpha \cong 200$ Å would indicate the maximum distance from the surface at which the alpha-emission would result in material ejection. Exceeding this distance, the recoil nucleus would produce considerable disruption without effecting, however, any material release from the particle immediately following the event.

Assuming a generalized $^{239}PuO_2$ particle, perfectly spherical, of a radius R, it would follow that alpha-disintegrations occurring at a radius $r \geq R - D_\alpha$ contribute to immediate mass ejection, whereas those at $r < R - D_\alpha$ would not. Consequently, the fraction of alpha-decays resulting in instantaneous mass release can be estimated on a volumetric basis from equation (37) (see also Figure 4).

$$f(R) = G\left[\frac{R^3 - (R - D_\alpha)^3}{R^3}\right] \qquad (37)$$

Where R = particle radius, μm

D_α = alpha-recoil nucleus range, μm

= .02 μm (Fleischer, 1975)

G = geometric factor, allowing for oblique recoils and for those moving inward rather than outwards

= 1/4 (Fleischer, 1975)

For particles of pure $^{239}PuO_2$ of 10 μm diameter, assumed to constitute the upper limit of resuspendible size, f(5 μm) ≅ 0.3 percent. At the lower limit, 1 μm diameter particles would give f(0.5 μm) ≅ .3 percent. If the particles of primary interest are those apt to be inhaled, ≤ 3 μm diameter (Stewart, 1967), then the fraction f(R) should vary between 1 percent (at r = 1.5 μm) to 3 percent (at r = 0.5 μm), with 2 percent assumed as an average.

The relevance of the preceding discussion can be best explained by describing a physical situation or "scenario" which the above processes may affect.

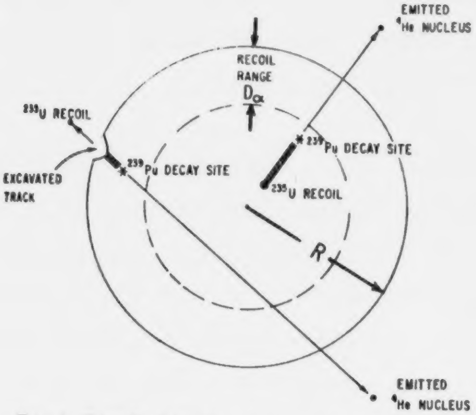

FIG. 4. Recoil ejection of atoms. Alpha decay of ^{239}Pu ejects the recoil ^{235}U nucleus if it was generated within its range D_α of the surface. A portion of the atoms along the track are ejected in the process.

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SCENARIO #1

1. A certain amount of activity was determined for a given soil upon deposition.

2. The resuspendible fraction of this activity originates from spherical particles of $^{239}PuO_2$ of physical diameters between 10 micrometer and 1 micrometer.

3. Following alpha-emissions, cones of n molecules of $^{239}PuO_2$ (containing n atoms of plutonium) are expelled from some of these spherical particles.

4. Because the emitted cone of material is considerably smaller than 1 micrometer, it attaches readily (and strongly) to some part of the soil surface.

5. Since the greater part of any soil is expected to move in creep and saltation but not in suspension, there is a strong probability that the ejected material is no longer available to resuspension. A quantitative indication of how small this probability is may be provided by Langham's resuspension factor of 10^{-6}.

6. In this case, the interest is in determining the resuspension availability of particles of hazardous size, $1\ \mu m \leq$ diameter $\leq 3\ \mu m$.

7. The surface concentration of such particles being N_H, μCi/m , the direct ejection model can be employed to describe the reduction in availability of hazardous particles, as per equation (38).

$$\frac{dN_H}{dt} = -\lambda f(R) n N_H \tag{38}$$

where N_H = surface concentration of particles of interest, atoms/m^2 or μCi/m^2.

λ = radioactive decay constant of plutonium-239,

= 7.813×10^{-8} days^{-1}

(R) = fraction of decays resulting in direct emission of n atoms of plutonium-239, dimensionless,

≈ 0.02 for range of particle sizes of present interest.

n = number of atoms dislodged per decay of atom of plutonium-239

≈ 3333 atoms of plutonium-239/decay

$$\frac{dN_H}{dt} = -5.21 \times 10^{-6} \text{ days}^{-1}\ N_H \qquad (39)$$

This, of course, is equivalent to an effective half-life of 133,061 days or 365 years, an imposing length of time which is, however, much shorter than the half-life of plutonium-239 itself, 14,300 years.

If instead of gaging the diminishing hazard of particles of respirable size, the interest were focused on the entire gamut of resuspendible sizes the corresponding value of f(R) would diminish accordingly. Assuming an average value of f(R) = 0.5 percent for the range of such sizes, the effective half-life would be over 500,000 days, or approximately 1,500 years.

Resorting to the alternative model of material removal mentioned by Fleischer, in which atomic disarray along the alpha-recoil tracks may lead to the elimination of 30-80 times the amount of atoms per event, the stage is set for a second presentation.

SCENARIO #2

Similar in most essentials to scenario #1. Significant alteration involves an increase by a factor of 80 in the number of atoms removed per event.

$$\frac{dN_H}{dt} = -\lambda f(R)(80r)N_H = -4.17 \times 10^{-4} N_H \qquad (40)$$

This would result in an effective half-life of 1663 days or 4.5 years. Using the lower limit of 30 in the above calculation would produce a half-life of approximately 12 years.

No credit was taken in the direct ejection model for disarray along the recoil nucleus track prior to cone ejection or for disruptions within the particle due to events not leading to ejection, but possibly to eventual fragmentation. Both these effects may lead to an increased similarity of the two models. In addition, perfectly spherical particles of pure $^{239}PuO_2$ were assumed as the source of radioactivity. That this assumption may be unwarranted can be best illustrated by a photograph used by Corn (1968) to emphasize a similar point (Figure 5). Thus, it appears that $^{239}PuO_2$ would not necessarily take the form of discrete spheres, but rather that of particles of undefined shape of which Figure 5 is a good, though possibly extreme, example. These somewhat abstract shapes would enhance the adhesion capabilities of the particle by virtue of greater surface area. Consequently particles that would resuspend readily, were they spherical, would in actuality remain attached to the soil or, if resuspended at all, to be found in close attachment to inactive material.

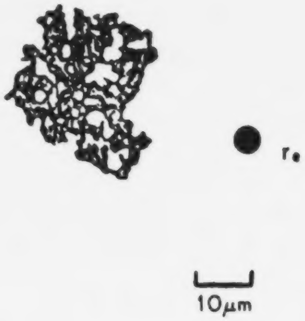

Figure 5. Atmospheric Particle and Sphere with Equivalent Aerodynamic Behavior. Reproduced from Stein, Quinlan and Corn, American Industrial Hygiene Association Journal. 27, 39 (1966), by permission.

As mentioned elsewhere, specks of plutonium that had settled on various surfaces were later resuspended as much larger particles, only partly composed of plutonium (Moss, Hyatt, Schulte, 1961). In a resuspension experiment conducted at Rocky Flats, particles of diameters up to seven micrometers were collected, attached to some of which were plutonium particles of sizes much smaller than the impactor size diameter of the collected fraction (Sehmel, 1975). Fission track analyses were conducted which established that all the plutonium present was in amounts corresponding to equivalent PuO_2 particles of diameters 0.06 micrometers to 0.25 micrometers (Sehmel and Lloyd, 1974; Sehmel, 1975). This is not meant to imply that "equivalent particles" of diameters less than 0.06 micrometers were not present; merely that they were not observed. As Moss et al. (1961) pointed out, the exposure time required to produce ten tracks per 0.01 micrometer $^{239}PuO_2$ particle would be roughly 50 years. Nor should the inference be drawn that the plutonium was actually present in spherical form. Although the technique used did not determine whether or not these particles were attached to larger host particles, the author of the present report would like to presume that they were, in agreement both with Chepil and the line of his own argument.

Assuming that the particles of $^{239}PuO_2$ were attached to other, larger particles, is tantamount to assuming that they were not spherical. If so, that would imply that the plutonium must be distributed along the surface of the host particle. Further, in order for adhesion to occur, plutonium would have to be present in small amounts. That would mean that the plutonium layer, where present, is thin. Consequently, the radius factor f(R) can now be ignored or set as $f(R) \approx 1$, which introduces a third scenario.

SCENARIO #3

1. The resuspendible fraction of the deposited activity is associated with particles of irregular shape to which plutonium has adhered in thin layers

2. Following an alpha-emission, n atoms of plutonium are emitted, subsequently becoming attached to indurated or non-resuspendible material

   n = 1/3 (800,000) atoms, as per disarray model

   f(R) = 1 [including geometric factor G = 1/2, but also a "break-up" factor = 2 (Fleischer, 1975)]

3. For a total concentration N of resuspendible material, atoms/m^2 or $\mu Ci/m^2$, the decrease in resuspension availability can be found from equations (41) and (42)

$$\frac{dN}{dt} = -\lambda n N = -0.0208 \text{ days}^{-1} \; N \tag{41}$$

or

$$N(t) = N(o)\, e^{-\frac{0.0208}{\text{days}} t}, \text{ t in days} \tag{42}$$

The factor 0.0208 days^{-1} is equivalent to a half-life of 33.3 days, remarkably close to the half-time empirically determined by Langham of 35 days. This result was obtained by deliberately selecting from the available information those conditions and assumptions that would, tentatively, lead to it. The result suggests the need for further study of the consequences of alpha-emissions in particulate matter, the effects of shape and size of deposited radioactive pollutants, and a survey of predominant shapes of plutonium dioxide particles both in pure PuO_2 form and as found in attachment to larger particles. The many countering arguments that may be found to the preceding discussion would only further emphasize the need.

Should the connection between radioactivity and decrease in availability of resuspendible material be more formally established, a model of such decrease could, when coupled with suitably quantified "ground penetration" and non-radioactive "weathering" models, be of some assistance in relating resuspended plutonium concentrations to soil type and conditions as well as type of release as classified by particle size and shape distributions.

8. ANSPAUGH'S MODEL

Langham's model does not include consideration of possible causes for decreased availability of resuspendible material. It is simply an empirical relationship, based on defining the time-dependent fraction of the total deposition which is available for resuspension coupled with a constant resuspension factor. Anspaugh et al. (1974, 1975) were of the opinion that such time-dependent fraction could not be realistically determined, and that it would be more advantageous to define a time-dependent resuspension factor assuming a constant value for the soil concentration, now equal to the total deposition per unit area, regardless of distribution with depth. Expressing Langham's and Kathren's models in this format,

$$C(t) = R_f(t)S_A \tag{43}$$

where $$R_f\,(t) = R_f(o)e^{-\lambda_A t} \tag{44}$$

with $C(t)$ = air concentration, μCi/m

$R_f\,(t)$ = time-dependent resuspension factor, m^{-1}

$R_f\,(o)$ = initial resuspension factor, at time $t = o^+$, m^{-1}

= $10^{-4}m^{-1}$ (Kathren, 1968)

= $10^{-6}m^{-1}$ (Langham, 1969a, 1971)

[Langham also reported an original value of $7x10^{-5}m^{-1}$ found by Langham and Harris, 1956.]

λ_A = attenuation constant, days^{-1}, equivalent to $T_{1/2}$ = 35 days (in Langham's model)

or $T_{1/2}$ = 45 days (in Kathren's model)

S_A = total soil activity per unit area, μCi/m^2

t = time, days

It should be mentioned, in passing, that Langham's original model was somewhat more general in applicability. Given an initial surface activity per unit area $S_a(o)$, which actually could be measured at any time after deposition, the model predicts an air concentration $C(t)$ at some time t *after this initial measurement*. Anspaugh's version of Kathren's and Langham's models, as it appears in equations (43) and (44), eliminates this flexibility, as can be proven by comparing the two versions predictions of air concentration at time t = o.

Langham's original model: $C(o) = R_F S_a(o)$ (45)

Anspaugh et alii version: $C(o) = R_f(o)\ S_A$ (46)

Note that unless $S_a(o)$ were measured at t = o = deposition time, at which time $S_a(o)$ and S_A would indeed be equal, R_F would be expected to be greater than $R_f(o)$ for air concentrations to be the same, since normally *surface* activity per unit area S_a can be expected to be less than *total* activity per unit area S_A. To preclude loss of flexibility $R_f(o)$ would have to have to be measured anew for each unspecified initial time, which clearly was not Anspaugh's intent, since he set $R_f(o) = R_F = 10^{-6}/m$(Figure 8). It follows that in doing so he restricted Langham's formulation to mean "t = o" as deposition time.

The relationships expressed by equations (43) and (44) approximate fairly closely the values observed for up to several weeks after the contaminating event. Anspaugh et al. (1975) quoted a number of observations that indicate they are inaccurate for longer periods of time:

1. A half-time of about 10 weeks was determined by Anspaugh et al. (1973) from observations made 12-40 weeks after release (accidental venting of an underground nuclear explosion, Figure 6).

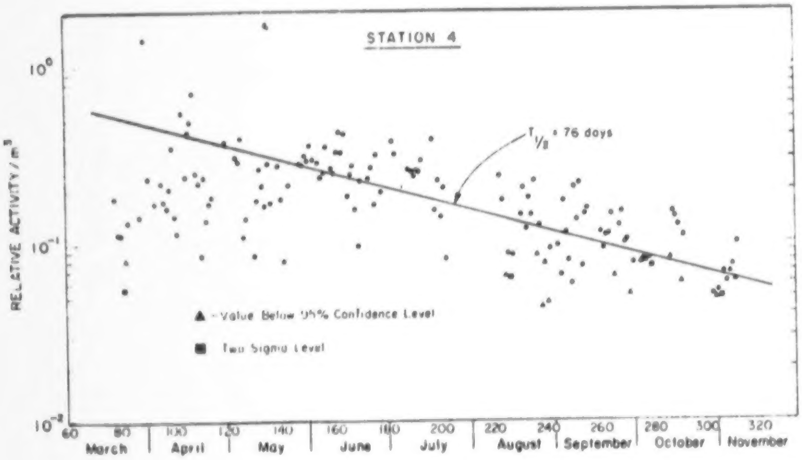

Figure 6. Least-squares fit to gross-gamma air activity levels three to eleven months following Baneberry venting. [from Anspaugh et al. (1973)]

2. A half-time of approximately 9 months was reported by Sehmel and Orgill (1974) after artificial disturbance (ditch digging) of a 10-year-old source.

3. A series of 236 individual measurements of air concentrations, at a location contaminated 17 years previously resulted in an average resuspension factor of $10^{-9}\,m^{-1}$. For the same time period, Langham's and Kathren's models as expressed by equations (43) and (44) would produce values of 10^{-60} and 10^{-46}, respectively.

To conform to these additional constraints, a simple model was proposed by Anspaugh et al., which can be represented by equation (47):

$$R_f(t) = R_f(o)e^{-\lambda \sqrt{t}} + R_f(\infty) \qquad (47)$$

where $R_f(t)$ = time-dependent resuspension factor, m^{-1}

$R_f(o)$ = initial resuspension factor, at $t = o$, m^{-1}
= $10^{-4}\,m^{-1}$

$R_f(\infty)$ = final resuspension factor, at $t = \infty$, m^{-1}

= $10^{-9}\,m^{-1}$

t = time, days

λ = $0.15/\sqrt{day}$

The term 10^{-9} was added on the assumption that there would be no measurable change after 17 years.

Replacing $\sqrt{t}$ with $t/\sqrt{t}$, it is easy to see that the exponent $(0.15/\sqrt{days})\sqrt{t}$ is equivalent to the expression $(0.15/\sqrt{days})(t/\sqrt{t})$. Isolating "t" in the preceding expression enabled the present author to define the remainder as a "time-dependent attenuation factor" $\lambda_a(t)$

$$\lambda_a(t) \equiv \frac{\lambda}{\sqrt{t}} = \frac{0.15/\sqrt{days}}{\sqrt{t}} \qquad (48)$$

Similarly, a "time-dependent half-time" $T_{1/2}$ was defined, based on $T_{1/2}(t) = \ln 2/\lambda_a(t)$ (figure 7),

$$T_{1/2}(t) = 4.62\sqrt{days}\ \sqrt{t} \qquad (49)$$

Anspaugh's model was derived empirically from results obtained at various locations having roughly similar terrain. It provides, as Anspaugh stated, "no fundamental understanding of the resuspension process," but intends merely to describe it. From Figures 7 and 8, the essential features of the model are evident. Nevertheless they are summarized below.

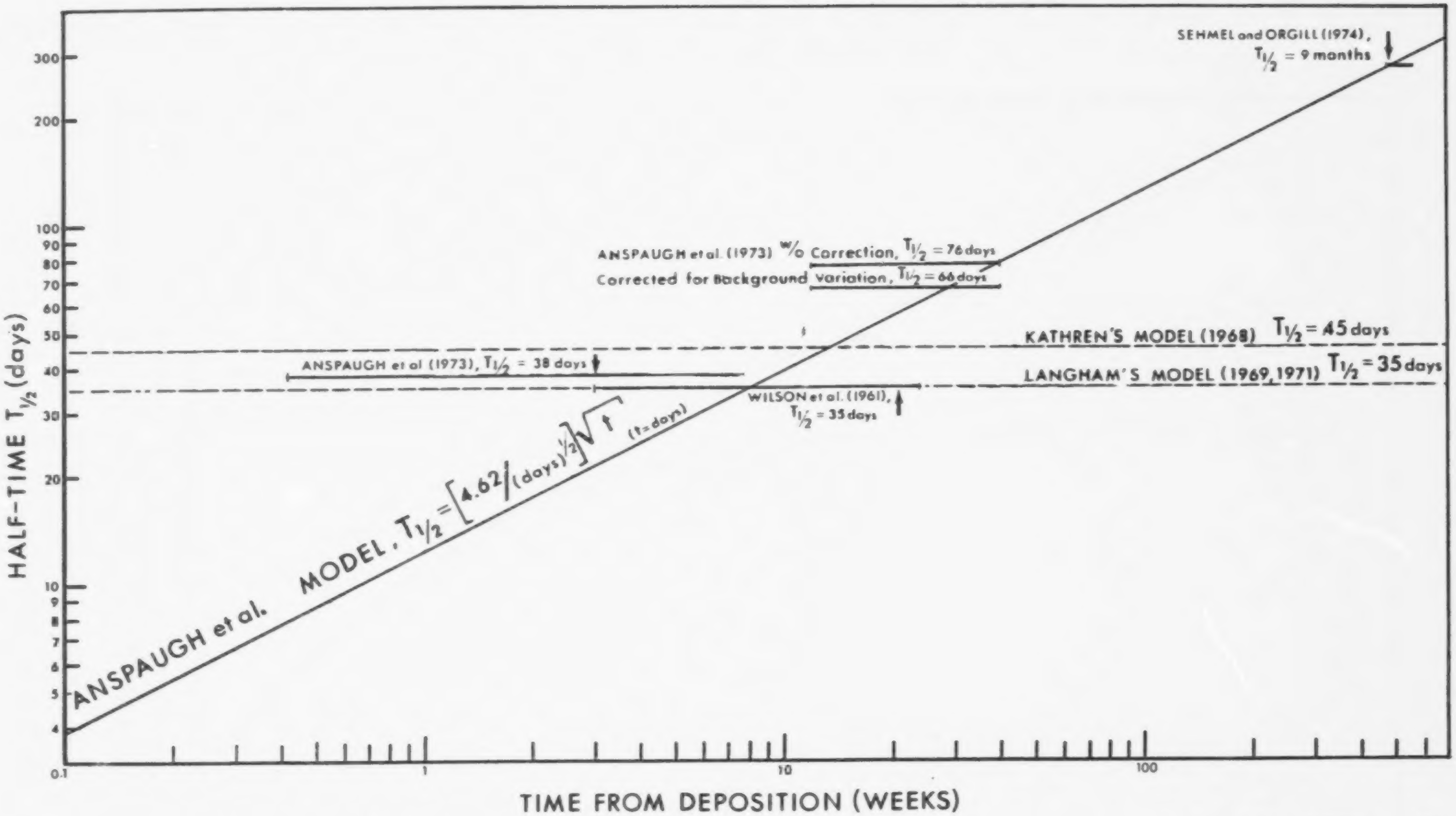

Figure 7. Time-dependent half-time, $T_{\frac{1}{2}}(t)$, resultant from exponent $(0.15/\sqrt{\text{days}})\ \sqrt{t}$ in Anspaugh's model, compared to experimentally determined half-time values

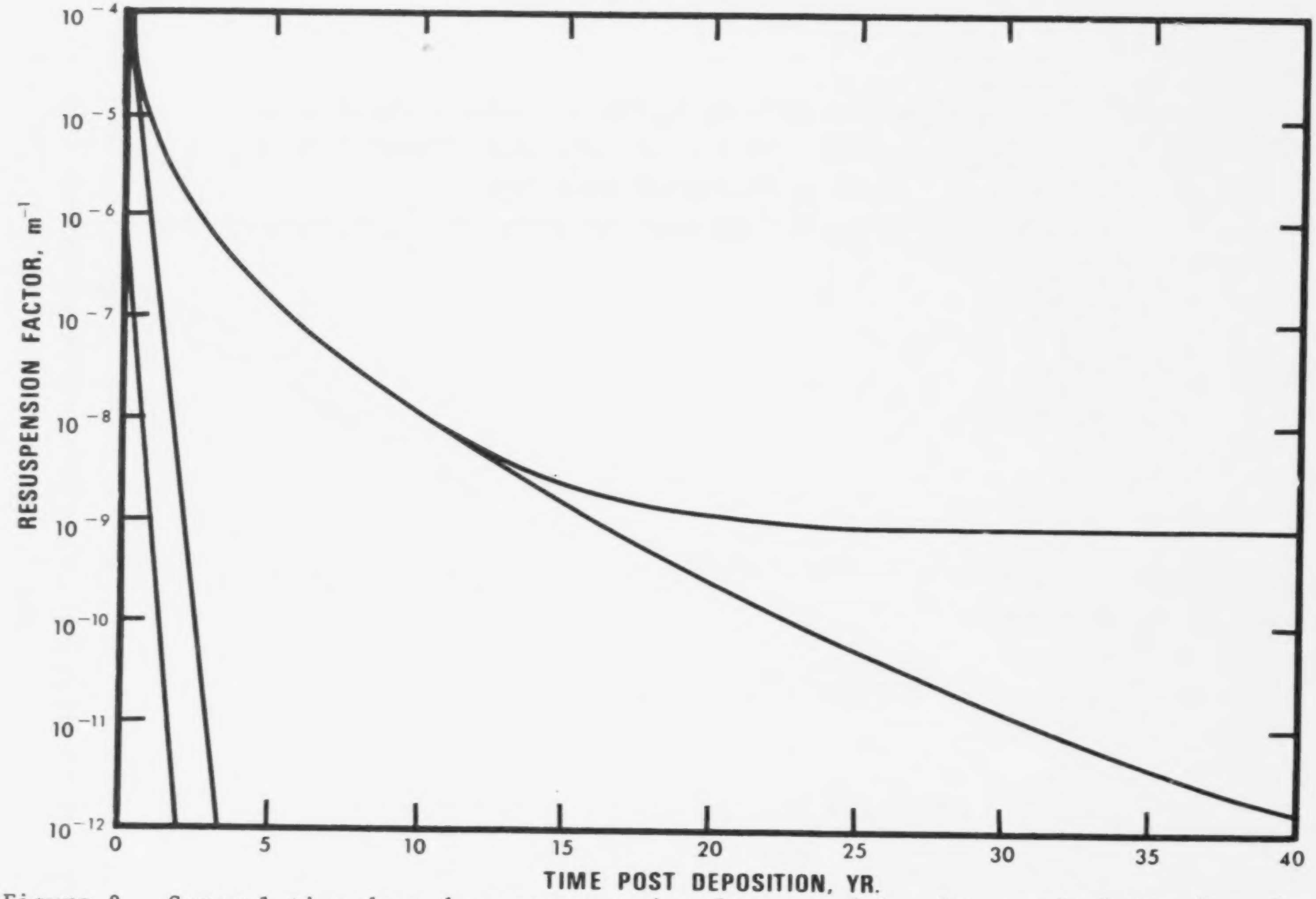

Figure 8. Several time-dependent resuspension factor models. Counterclockwise from far left: Langham's, Kathren's, Anspaugh's model without the constant term $10^{-9} m^{-1}$, Anspaugh's model with the constant term [After Anspaugh et al. (1974)]

1. at $t \cong 8$ weeks, $T_{1/2} = 35$ days
2. at $t \cong 33$ weeks, $T_{1/2} = 70$ days
3. at $t \cong 10$ years, $T_{1/2} \approx 9$ months
4. at $t \geq 25$ years, $R_f = 10^{-9}\,m^{-1}$

Beginning with a value of $10^{-4}\,m^{-1}$ at $t = o$, the model shows a drop to half this value in approximately three weeks, to a quarter of it in an additional 9 weeks, to an eighth in another 18 weeks, proceeding in this fashion until values comparable to $10^{-9}\,m^{-1}$ are reached. At that point, the presence of this constant term forces the model into a different type of behavior, i.e., an asymptotic approach to the value $10^{-9}\,m^{-1}$. Obviously the relationships described by equations (48) and (49) no longer hold in this region of time.

Note that Anspaugh's model, unlike Langham's or Kathren's, makes reference to a specific point in time, $t = o$, from which events take their course. Consequently, the model portrays resuspension as after an "instantaneous" release of pollutant (detonation) or a release of short duration. A continuous release is unlikely to provide the researcher any such definite point in time.

Following an explosive release, the airborne concentrations of radioactive aerosols rapidly decrease by factors of 100 to 1000 in some 100 hours (Anspaugh, 1975). After this initial rapid decrease, slower rates of decline are apparent, in a manner roughly similar to that modeled by Langham's, Kathren's, and Anspaugh's resuspension factors (Figure 9). Note that Anspaugh's model predicts, at 1000 hours after $t = o$, a resuspension factor of 0.378 times the initial value at $t = o$. Air concentrations follow suit. Obviously, Anspaugh's model does not show the initial rapid drop, during which, presumably, deposition is still in process. The present author interprets this to mean that an air concentration 100-1000 times larger than that resultant from

Anspaugh's expression should be expected, as a first approximation, roughly 4 days (100 hours) before $t_m = 0$ in the model (where the subscript "m" indicates "model initial time," equivalent to t = 100 hours after the actual release).

It may be of interest to compare these predicted relationships to actual measurements obtained in Project Schooner, which influenced the model. Continuing to assume that $t_m = 0$ in the model is equivalent to t = 100 hours after detonation, Figure 10 shows the expected ratios graphically, with the air concentration at t = 100 hours ($t_m = 0$) equal to 10^{-3} that obtained at $t = 10^{-1}$ hour, and 0.378 of the 100-hour concentration to be found at t = 1100 hours ($t_m = 1000$ hours). Using the graph for station 11 as an example, the air concentration at $t = 10^{-1}$ hour (marked with a cross) is seen to be approximately 5.5×10^5 pCi/m^3. The corresponding concentration at t = 100 ($t_m = 0$) must be 5.5×10^2 pCi/m^3, and that at t = 1100 hours ($t_m = 1000$ hours) should be 2.08×10^2 pCi/m^3 (similarly marked). The same procedure was followed for the other stations measurements, with exception of station 23, which was excluded from consideration in determining resuspension factors by Anspaugh et al. (1970).

Examining Figure 10 , it appears that the 100-hour values obtained from the above described exercise, at stations 5, 11, and 29, are quite plausible, inasmuch as they seemingly fall on an imaginary smooth curve describing the overall behavior of the air concentration at these stations. The values at 1100 hours at these same stations, however, exceed the expected "reasonable" values (indicated by circles) by factors of two or three. This suggests immediately that, if the model $t_m = 0$ occurs, indeed, at 100 hours after detonation, both the initial value of the resuspension factor and the initial rates of change should be somewhat higher if the model is expected to accurately portray the behavior of air concentrations for $t \geq 100$ hours after detonation.

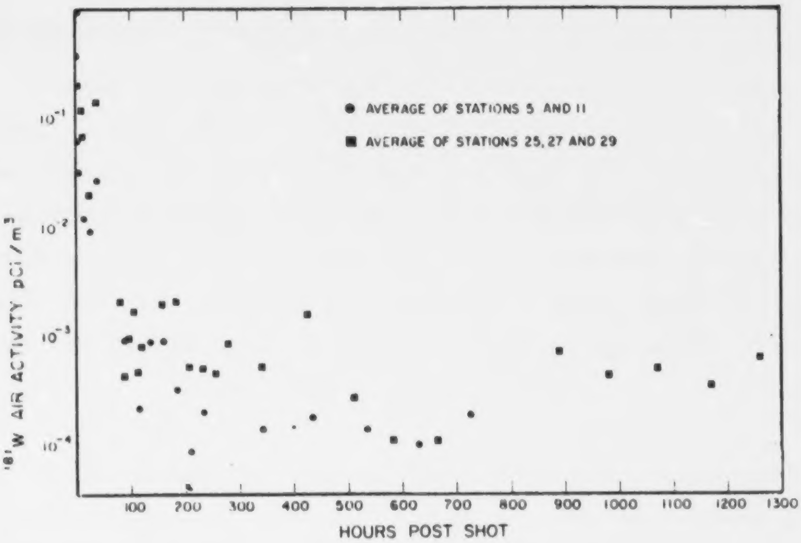

Figure 9. Tungsten-181 air activity as a function of time, from Project Schooner. [From Anspaugh et al. (1973)]

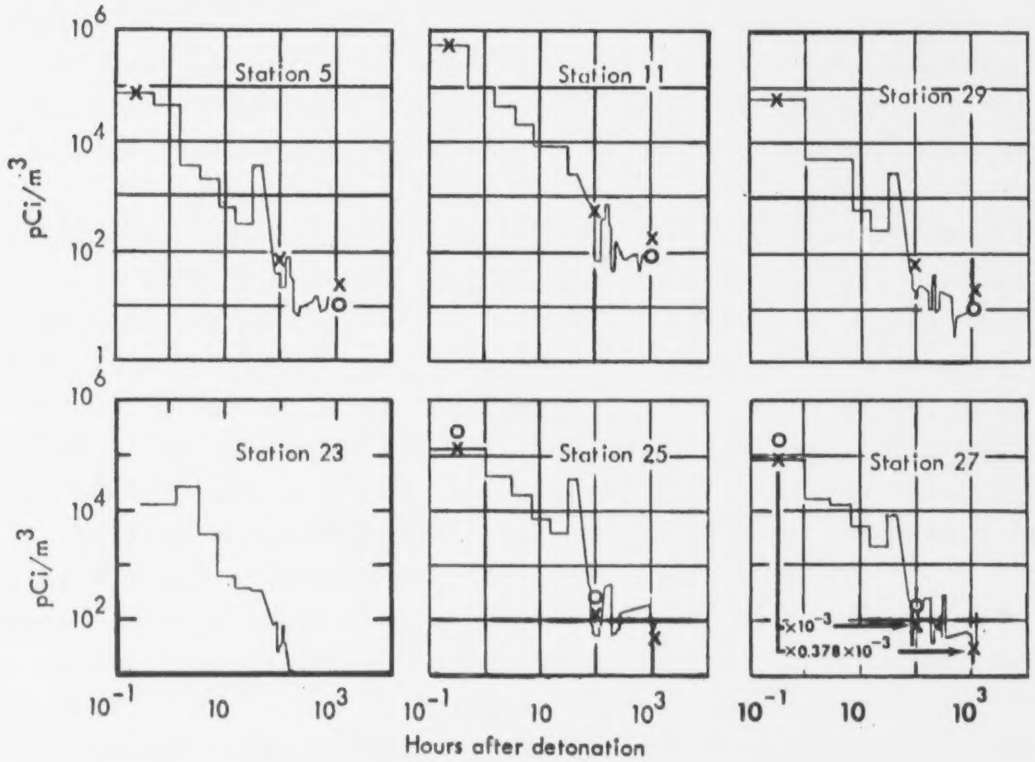

Figure 10. Tungsten-181 air activity levels (corrected to zero time) as a function of time at six stations, from Project Schooner [Adapted from Anspaugh et al. (1970)]

The reverse situation exists at stations 25 and 27. There the model values at t = 1100 hours (t_m = 1000 hours) seem to agree closely with the overall behavior of the graphs, whereas the values at 100 hours do not. Raising these values by factors of two or three, however, provide the modeler with a better fit, although this also raises the initial values, at 10^{-1} hour (shown by circles). This reinforces the point discussed in the previous paragraph.

The import of this exercise is not to demonstrate a divergence by a factor of two or three between the expected initial value of resuspension factor and that provided by the model; but to point out that, if the resuspension factor at t_m = 1000 hours (in the model) is indeed supposed to be ≅ 0.378×10^{-4}, then the initial value of 10^{-4} is not by any means a conservative upper limit, since it can easily be exceeded in order to provide better agreement with data. The values of $10^{-4} m^{-1}$ and $10^{-6} m^{-1}$ used by Kathren and Langham, respectively, were not intended for use as initial values for a time-dependent resuspension factor, but rather as constant values expected to apply to a number of diverse conditions and times after deposition. Thus, inevitably, they must be average values. Langham, in fact, felt that "intuitively,... a factor of about 10^{-6} is a reasonable average value to use in estimating the potential hazard of occupancy of a plutonium contaminated area; however, intuition is not a convincing argument." Previously, he had obtained values of ≅ $7 \times 10^{-5} m^{-1}$ "at two different times after the event" for disturbed Nevada desert conditions (resulting from extensive vehicular traffic) and ≅ $7 \times 10^{-6} m^{-1}$ from equilibrium calculations with dusty rural air. Kathren chose $10^{-4} m^{-1}$ as a conservative value "satisfactory at this time" due to absence of adequate data, although he made mention of "resuspension factors as great as $7 \times 10^{-4} m^{-1}$" from weapons experiments with both plutonium and uranium. Again, since Kathren's $10^{-4} m^{-1}$ value was intended for use at any time after deposition, it may well be said that it is "conservative."

It does not follow that it would necessarily be so "conservative" as an initial value. A factor of $10^{-3}m^{-1}$ would appear to be a more realistic upper limit for initial values of the resuspension factor. Note that values of $R_f = 10^{-3}m^{-1}$ are not entirely unknown in the literature. Table 5 (Stewart, 1967) points to comparable values having been measured previously, though not necessarily as initial values, which could, presumably, be even greater. Langham mentioned a value of $R_f = 10^{-3}$ being used by a member of the Danish scientific team during the Thule deliberations (Langham, 1969b).

In the context of the preceding discussion, it should be made clear that the use of $t_m = 0$ at $t = 100$ after release is the author's interpretation, based on examination of Figures 9 and 10. Anspaugh et al. did not specify a "model initial time," as suggested by Figure 8, which shows R_f (t) as function of time "post deposition," not "post release." However, Anspaugh et al. (1970) pointed out that cloud passage did not result in abrupt changes in air activity, and interpreted this to mean that resuspension and redistribution start "immediately," gradual loss of activity being due to weathering rather than to cloud passage. Assuming that "immediately" means "at release" ($t = t_m = 0$), the model rates of change between $t = t_m = 100$ hours and $t = t_m = 1100$ hours should be even slightly smaller than those predicted with the exercise in preceding paragraphs, and the discrepancy with the observed concentrations in Figure 10 slightly greater. In addition, equating $t = t_m = 0$ would make it extremely difficult to support any estimates of an initial value and rate of change for R_f, since researchers do not have, presumably, the ability to readily distinguish between "resuspended" contamination and that contained in the original "release" cloud.

In conclusion, assigning an initial value to R_f (t) appears to be premature on the basis of presently available data, unless $t_m = 0$ (deposition time) is specifically defined to represent a

TABLE 5. SHORT SUMMARY OF EXPERIMENTAL RESULTS ON RESUSPENSION OF ACTIVITY IN THE AIR [After Stewart (1967)]

Measurement Conditions	Resuspension Factor, R_F (m^{-1})	
	Range	Mean
Plutonium sampled at 1 ft above ground (1)		
Vehicle traffic	$3x10^{-4}$ to $7x10^{-4}$	
Pedestrian traffic	$3.5x10^{-6}$ to $3x10^{-4}$	
Particle size: Mainly 20-60 μm, with 1% in hazardous range (∿ 3 μm for PuO_2)		
Uranium sampled downwind from a crater (1)		
At 1 ft. above ground(dust stirred up)		1×10^{-3}
At 1 ft. above ground		3×10^{-4}
At 2 ft. above ground		1×10^{-5}
Brick/plaster dust sample contaminated with I-131 (2)		
Enclosed space	$2x10^{-4}$to $4x10^{-5}$	
Open space		$2x10^{-6}$
Sample in cab of Landrover, after a test (1)		
Round 1 (H + 18 hr)		2.5×10^{-5}
Round 2 (H + 5 hr)		6.4×10^{-5}
Airborne material without artificial disturbance of ground, consisting of limestone rock and sand with coarse grass and small bushes (3)	$1x10^{-6}$ to $8x10^{-5}$ (12 results)	1×10^{-5}
Random samples following a tower shot, without artificial disturbance, near crater (3)	$1x10^{-6}$ to $1x10^{-8}$ (9 results)	2×10^{-7}
On two roads formed by soil grading -no artificial disturbance (3)	$1.5x10^{-6}$ to $1x10^{-6}$ (14 results)	2.5×10^{-7}
At back of a moving Landrover (3):		
D-Day + 4 (21 results)	$8x10^{-7}$ to $3x10^{-5}$	1.4×10^{-5}
D-Day + 7 (21 results)	$6x10^{-7}$ to $4x10^{-6}$	1.5×10^{-6}
D-Day + 7 over tailboard	1.6 and $3.1x10^{-5}$	2.5×10^{-5}

---

(1) From nuclear weapon and other tests at Maralinga
(2) From Civil Defense trial at Falfield, Gloucester
(3) From Hurricane Trial

time t = x hours or days after release at which actual measurements of R_f have been made.

9. SHORT CRITIQUE OF RESUSPENSION FACTOR CONCEPT

Having discussed some possible shortcomings of Anspaugh's, Langham's and, by extension, Kathren's time-dependent resuspension models, some serious criticism can be leveled at the concept of resuspension factor in general. In the first place, it assumes that the air concentration above a contaminated surface is directly proportional to this surface contamination level, rather than on the extent of ground contamination upwind of the sampling site, as would be more logical and, in fact, was found to be true (Stewart, 1967; Mishima, 1969). In the second place, the constant of proportionality relating air concentration to ground contamination,(i.e., the resuspension factor) is meant to include the effects of a myriad of parameters, such as wind velocity, surface roughness, physical and chemical characteristics of the soil surface, vegetation cover, etc. Whereas some of these factors may change little at a particular site, some others, such as wind velocity, tend to be less constant. This should justify the conclusion that the resuspension factor is an empirically determined value applying only to prevailing conditions at a given site, at a given time. Thus, it is not surprising that the measured values of R_f vary from 10^{-2} to 10^{-13} m^{-1}, as shown in Table 6, (Mishima 1964). Therefore, even if average wind velocities are considered to be site-dependent variables (thus apt to be incorporated into the R_f for a given site), and the surface contamination is assumed to be fairly uniform over a large area and constant with time (consequently minimizing the difference between "upwind" and "local" effects), the fact remains that the resuspension factor is of limited value in predicting air concentrations at any site other than one for which the R_f has been found (via previous measurements of air and ground concentrations). Even at such a site, seasonal variations,

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TABLE 6. RESUSPENSION FACTORS FOR PLUTONIUM AND OTHER RADIOISOTOPES [FROM MISHIMA (1964)]

Conditions of Resuspension	Resuspension Factor
Average Resuspension Factor in Accidents Involving Plutonium	4×10^{-6} m^{-1}
Vehicular Traffic (Nevada)	7×10^{-5}
People Working or Active in a Closed Area	4×10^{-5}
Dirty Rural, Suburban, or Metropolitan Areas	7×10^{-6}
People Working or Active in an Open Area	2×10^{-6}
Isolated Area	$\sim 7 \times 10^{-7}$
Resuspension of Aged Plutonium Deposit (0.74 to 752 μCi/m^2) from "Plumbob"	6.2×10^{-10} to 10^{-13}
Plutonium Oxide, No Movement	2×10^{-8}
Plutonium Oxide, 14 steps/min	10^{-5}
Plutonium Oxide, 36 steps/min	5×10^{-5}
Plutonium Nitrate, No Movement	2×10^{-8}
Plutonium Nitrate, 14 steps/min	10^{-6}
Plutonium Nitrate, 36 steps/min	5×10^{-6}
Plutonium Oxide, Change Room (>3000 ft^2), 9 air changes/hr, 0.01 μCi/m^2, 4 to 6 persons active in area:	
"Loose" Contamination (estimated by smears)	10^{-3}
"Loose" Contamination (estimated by water-detergent wash)	2×10^{-4}
Changing Coveralls, Static Sampler, No Ventilation	2.8×10^{-3}
Changing Coveralls, Personal Sampler, No Ventilation	6.4×10^{-3}
Personnel Traffic in a Small Unventilated Room	4×10^{-3}
Proposed Resuspension Factors for Plutonium Oxide:	
Outdoors (quiescent conditions)	10^{-6}
Outdoors (moderate activity)	10^{-5}
Indoors (quiescent conditions)	10^{-6}
Indoors (moderate activity)	10^{-4} to 10^{-5}
From Crater of Tower Shot, No Artificial Disturbance	8×10^{-8}*
Survey of Road, No Artificial Disturbance	8×10^{-8}*
Survey of Road, Landrover, D-Day + 4	1.4×10^{-5}
Survey of Road, Landrover, D-Day + 7	1.5×10^{-6}
Survey of Road, Tailboard of Landrover, D-Day + 7	2×10^{-5}
Survey of Road, D-Day + 1 and 2	4×10^{-7}
Sample Collection in Cab of Landrover, H-Hour + 5	6.4×10^{-5}**
Sample Collection in Cab of Landrover, H-Hour + 8	2.5×10^{-5}†
Uranium Sample Downwind of Crater, Sample Height: 1 ft Above Ground	3×10^{-4}††
Uranium Sample Downwind of Crater, Dust Stirred Up, Sample Height: 1 ft	10^{-3}††
Uranium Sample Downwind of Crater, Sample Height: 3 ft	10^{-5}††
Plutonium Sampled 1 ft Above Ground:	
Vehicular Dust	5×10^{-4}§
Pedestrian Dust	1.5×10^{-6}§
Iodine-131, Enclosed (Chamberlain & Stanbury)	2×10^{-4} to 4×10^{-5}
Iodine-131, Open (Chamberlain & Stanbury)	2×10^{-6}
Yttrium-91, 0-8 μ Particles, Natural Turbulence, Sampled 1 ft Above Ground:	
Ground Contamination Level 1.8 μCi/m^2	1.8×10^{-7}
Ground Contamination Level 6.8 μCi/m^2	10^{-8}
Ground Contamination Level 24.6 μCi/m^2	3×10^{-10}
Polonium-210, 0-8 μ Particles, Natural Turbulence, Sampled 1 ft Above Ground:	
Ground Contamination Level 0.6 μCi/m^2	2×10^{-6}
Ground Contamination Level 5 μCi/m^2	2×10^{-6}
U_3O_8, 0-4 μ Particles, Natural Turbulence, Sampled 1 ft Above Ground:	
Ground Contamination Level 112 g/m^2	2.4×10^{-6}
Grass Contamination Level 70 g/m^2	8×10^{-6}
Concrete Contamination Level 180 g/m^2	2×10^{-6}
Plutonium Oxide, Sampling Height: 5 ft	
Floor Level 0.1 μCi/m^2, No Circulation	1.6×10^{-6}
Floor Level 24.6 μCi/m^2, No Circulation	4.4×10^{-7}
Floor Level 0.1 μCi/m^2, Fan	1.3×10^{-3}
Floor Level 0.91 μCi/m^2, Fan	1.4×10^{-3}
Floor Level 0.086 μCi/m^2, Fan and Dolly	1.0×10^{-2}
Floor Level 1.3 μCi/m^2, Fan and Dolly	9.4×10^{-3}
Floor Level 1.3 μCi/m^2, After Test	9×10^{-4}
Floor Level 1.1 μCi/m^2, After Test	9×10^{-4}
Uranium, Sampling Height: 5 ft	
Floor Level 0.086 μCi/m^2, No Circulation	1.8×10^{-6}
Floor Level 0.95 μCi/m^2, No Circulation	2.2×10^{-7}
Floor Level 0.095 μCi/m^2, Fan	1.5×10^{-4}
Floor Level 1.1 μCi/m^2, Fan	1.1×10^{-4}
Floor Level 0.11 μCi/m^2, Dolly	1.6×10^{-4}
Floor Level 1.3 μCi/m^2, Dolly	1.3×10^{-4}
Floor Level 0.91 μCi/m^2, Fan and Dolly	4.6×10^{-4}
Floor Level 1.0 μCi/m^2, Fan and Dolly	1.9×10^{-4}

* One high value excluded

† Only ~20% of particles <6 μ

§ Particles primarily in 20 to 60 μ size range, <1% <6 μ

** Only ~10% of particles <6 μ

†† Only ~5% of particles <6 μ

at least, should be expected. The limitations of the resuspension factor being well understood, alternative methods were considered, particularly in regard to what is undoubtedly one of the most important variables affecting resuspension, that is, wind velocity. These methods will be treated in the following sections.

10. SLINN'S MODEL

On the basis of the discussion in previous sections, it would appear that a general functional relationship defining resuspension factors in terms of wind velocity as well as other influencing variables is badly needed. One such formulation was provided by Slinn (1975) as

$$R_f = \frac{3.5 \times 10^{-9}\,\mathrm{cm/sec}}{1\ \mathrm{ton/acre.month}}\left(\frac{rE}{2\pi uS}\right) \tag{50}$$

Where

$\dot{E} = f(I,R,C,V,L)$ in ton/acre.month, soil erodibility, function of
- I = soil erodibility index,
- R = soil ridge roughness factor,
- C = climatic factor,
- V = equivalent quantity of vegetative cover,
- L = field length along prevailing wind direction,
- r = fraction of the horizontal flux at the height of the sampler, dimensionless. This can be estimated from

  $c = c_o\,(Zo/_Z)^{0.28}$

  where:
  - c = weight of dust in unit volume of air at height Z
  - c_o = function of I = soil erodibility index
    u = wind velocity, etc.

u = mean wind speed at sample height, cm/sec

S = depth to which the contaminant has penetrated the soil, m.

The effectiveness of this formulation in predicting resuspension factors and/or airborne concentrations was not reported. The present author assumes that, since soil penetration "S" is generally a very slow function of time, the overall decrease in R_f as time progresses would have to be linked primarily to a similar behavior in the soil erodibility. However, in at least some cases involving sandy soils the erodibility is supposed to increase with time, rather than the opposite (Chepil, 1957b). Thus it may be expected that, in general, Slinn's resuspension factor will not account for the "weathering effect" discussed in connection with radionuclide particles.

MASS LOADING

The resuspension factor approach is based on the implicit assumption that a state of equilibrium between resuspension and deposition processes has been achieved locally and further, by being applied over large areas, minimizes the effects of wind transport and saltation buildup observed downwind of a deposited source of contaminant.

One additional assumption, that of activity per gram of soil being equal to activity per gram of dust in the atmosphere and proportional to its activity per unit volume, leads directly to the "mass loading approach." The constant of proportionality, called "average mass loading of the atmosphere," in units of mass per volume, is defined for a particular land use and for a set of environmental conditions. As an example, if the average mass loading is estimated to be 100 $\mu g/m^3$ at a given site, and the specific activity per unit mass of soil is 3 nCi/g, the expected air concentration of pollutant can be found from:

$$C = AML \times A \tag{51}$$

Where C = air concentration, nCi/m^3
AML = average air mass loading, $\mu g/m^3$
A = specific ground activity, nCi/g

In this example,

$$C = 100\ \frac{\mu g}{m^3} \times 3 \times 10^{-6}\ \frac{nCi}{\mu g} = 3 \times 10^{-4} \frac{nCi}{m^3}$$

$$= 3 \times 10^{-13}\ \frac{\mu Ci}{cm^3}$$

This simple but effective model was employed by Anspaugh to predict air concentrations at a number of sites. Predicted values did not exceed measured values by more than a factor of roughly five in one extreme case (Anspaugh, 1975). The fallacy of this model is in assuming that the resuspendible fraction of the soil would carry with it an equal fraction of the activity, which implies essentially that 1) activity is distributed homogeneously in the top soil and, 2) activity exists independently of particle size. For instance, if the specific ground activity is associated mostly with particles of size greater than 50 micrometer, a very small air concentration would result, although the model would predict the same air concentration for this case as it would for all the activity being distributed among particles of resuspendible size. In either case the model would fail. It would seem reasonable to assume, however, that the error would be least for soils in which the contaminant has been present for some time (aged source). Table 7 shows some results.

SEHMEL AND ORGILL MODEL

It is axiomatic that any approach that ignores the physical conditions and processes affecting resuspension will be severely limited in its predictive ability. Recognizing wind speed as one of the most important parameters involved, Sehmel and Orgill (1973) attempted establishing a correlation between it and airborne concentration. Using the physics of soil erosion as a point of departure, more specifically that the rate of soil erosion is proportional to the air velocity cubed, it could be

TABLE 7. OBSERVED AIR CONCENTRATIONS COMPARED WITH CONCENTRATIONS PREDICTED BY MASS LOADING MODEL [ADAPTED FROM ANSPAUGH ET AL. (1974) AND ANSPAUGH ET AL. (1975)]

Location, etc.	Radionuclide	Air Concentration	
		Predicted[a]	Measured[b]
GMX site, USAEC Nevada Test Site [1]			
NE, 1971-1972	^{239}Pu	7200 aCi/m^3	6600 aCi/m^3
GZ, 1972, 2 weeks	^{239}Pu	120 fCi/m^3	23 fCi/m^3
Lawrence Livermore Laboratory			
1971 [2]	^{238}U	150 pg/m^3	52 pg/m^3
1972 [3]	^{238}U	150 pg/m^3	100 pg/m^3
1973 [4]	^{238}U	150 pg/m^3	86 pg/m^3
1973 [4]	^{40}K	1000 aCi/m^3	980 aci/m^3
Argonne National Laboratory [5]			
1972	^{232}Th	320 pg/m^3	240 pg/m^3
1972	^{nat}U	215 pg/m^3	170 pg/m^3
Sutton, England [6]			
1967-1968	^{nat}U	110 pg/m^3	62 pg/m^3

[a] Predicted value is equal to the soil concentration (activity/g) x 10^{-4} g/m^3.

[b] Most values are annual averages.

[1] ANSPAUGH, L.R., PHELPS, P.L., KENNEDY, N.C., BOOTH, H.G., GOLUBA, R.W., REICHMAN, J.M., KOVAL, J.S., Resuspension element status report, The Dynamics of Plutonium in Desert Environments, USAEC Rep. NVO-142 (1974).

[2] GUDIKSEN, P.H., LINDEKEN, C.L., GATROUSIS, C., ANSPAUGH, L.R., Environmental Levels of Radioactivity in the Vicinity of the Lawrence Livermore Laboratory, January through December 1971, USAEC Rep. UCRL-51242 (1972).

[3] GUDIKSEN, P.H., LINDEKEN, C.L., MEADOWS, J.W., HAMBY, K.O., Environmental Levels of Radioactivity in the Vicinity of the Lawrence Livermore Laboratory, 1972 Annual Report, USAEC Rep. UCRL-51333 (1973).

[4] SILVER, W.J., LINDEKEN, C.L., MEADOWS, J.W., HUTCHIN, W.H., MCINTYRE, D.R., Environmental Levels of Radioactivity in the Vicinity of the Lawrence Livermore Laboratory, 1973 Annual Report, USAEC Rep. UCRL-51547 (1974).

[5] SEDLET, J., GOLCHERT, N.W., DUFFY, T.L., Environmental Monitoring at Argonne National Laboratory, Annual Report for 1972, USAEC Rep. ANL-8007 (1973).

[6] HAMILTON, E.I., The concentration of uranium in air from contrasted natural environments, Health Phys. 19 (1970) 511.

assumed that resuspension is also a function of air velocity, to an unknown power. From diffusion theory,

$$\frac{C}{Q} = \frac{1}{u} f(\sigma,x,y,z) \tag{52}$$

where C = air concentration, μCi/m^3

Q = resuspension rate, μCi/hour

σ = standard deviation of distribution, m

x,y,z = space variables

u = average wind velocity m/sec, mph, etc.

As a first approximation, the particle resuspension rate Q, is assumed to be proportional to u^3. Thus,

$$C = \frac{u^3}{u} g(\sigma,x,y,z) \tag{53}$$

$$\text{or } C = u^2\ g(\sigma,x,y,z) \tag{54}$$

Experimentally, Sehmel found air concentration at sampling station S-8 at Rocky Flats, for one-hour periods, expressed by

$$C = 0.45\ (u)^{2.1} \tag{55}$$

where C = average airborne concentration, fCi/m^3

u = average wind velocity, miles/hour.

It is easy to see that Sehmel's empirical relationship is roughly consistent with the theoretical relationship predicted by diffusion theory. Similar analyses at other stations and for all time periods should determine whether this model is generally effective, and whether or not there is a minimum wind speed below which the model is unoperational. With the above simple model, the threshold velocity was found to be not significantly different from zero, at station S-8.

Neither the sources nor the ground concentrations were sufficiently defined for establishing a correlation between deposited and resuspended plutonium. A local activity of 0.3 fCi/m³ not directly attributable to the west and southwest winds used in developing the model was observed. However, maximum resuspension factors at Rocky Flats varied between 10^{-5} to 10^{-9} m^{-1}, for different time periods and different sites, which complicated possible analysis. Thus the model appears to be of somewhat limited applicability.

GILLETTE AND SHINN MODEL

Pursuing the relationship between resuspension and soil erosion, Shinn et al. (1974) developed a model relating dust concentrations and vertical aerosol fluxes to wind velocities and conditions of the soil.

The vertical flux of aerosols, assumed constant to within a few tens of meters from the ground, was expressed by Gillette et al. (1972) as

$$F_A = -K_A \rho \frac{\partial n}{\partial z} \qquad (56)$$

where F_A = vertical aerosol flux, positive when described upwards, in particles per unit area per unit time

K_A = coefficient of exchange for aerosols, in units of area per unit time

ρ = density of air, in mass per unit volume

n = number of particles per unit mass of air

Z = height, in length units.

Because the researchers' interest was limited to particles of less than 20 micrometer in diameter, the terminal velocity was much less than vertical velocity fluctuations, and the coefficient of exchange K_A was assumed to be, approximately $K_A = K$, where K is the eddy viscosity, and

$$K = u_* kz \tag{57}$$

where u_* = friction velocity, in units of length per time

k = 0.4, Von Karman's constant

z = height

Further, equation (56) was modified to represent mass, rather than number of particles, per unit area per unit time.

$$F = -K \frac{dC}{dz} \tag{58}$$

where F = vertical aerosol flux, mass per unit area per unit time.

C = concentration of aerosol mass per unit volume of air,

and thus equation (59) may now be written, from equation (57) and (58), as

$$F = -u_* kZ \frac{dC}{dz} \tag{59}$$

From dust profiles obtained at GMX and Texas and observations made in Kansas and Colorado, a power-law distribution with height was found for the dust concentrations. The exponents were either -0.25 or -0.35, with a value of -0.3 assumed for the model. Thus,

$$\frac{dC}{dz} = p \frac{C}{Z} \tag{60}$$

where $p = 0.3$

Combining expressions (59) and (60) and the value of k from equation (57), results in

$$F \cong 0.12\, u_* C \tag{61}$$

Since in the range of 0.7 m to 2.0 m the concentration varies only ~ ± 20 percent from the reference concentration C_1, at one meter, it was found convenient to specify flux, also, at this height, F_1, as follows:

1. Shinn recognized the importance of saltation flow in determining concentration, and also that saltation, friction velocity, and surface characteristics are interrelated. Empirical relationships were derived from observations at GMX and in Texas, of the form

$$C_1 = a\ u_*^{\gamma} \tag{62}$$

GMX $$C_1 = 6.1\ u_*^{2.09} \tag{63}$$

$$a = 6.1$$

$$\gamma = 2.09$$

Texas $$C_1 = 522\ u_*^{6.38} \tag{64}$$

$$a = 522$$

$$\gamma = 6.38$$

where C_1 = concentration at 1-meter, mg/m³

u_* = friction velocity, m/sec.

2. Applying the above relationships to equation (61), the fluxes at these locations can be expressed as

$$F_1 = b\ u_*^{\gamma+1} \tag{65}$$

GMX $$F_1 = 0.73\ u_*^{3.09} \tag{66}$$

Texas $$F_1 = 62.64\ u_*^{7.38} \tag{67}$$

In other words, concentration and flux can be expressed in the general form,

$$C_1 = a\ u_*^{\gamma} \tag{68}$$

$$F_1 = b\ u_*^{\gamma+1} \tag{69}$$

where $$b = 0.12a \tag{70}$$

The subscript "1" indicates indicates "at one meter above ground" By choosing $u_* = u_o = 1$ m/sec a convenient designation was found for the coefficient b, that of "reference dust flux"

$$F_o = b\ u_o^{\gamma+1} \tag{71}$$

where $u_o = 1$ meter/sec

or $$b = F_o/(u_o^{\gamma+1}) \tag{72}$$

Thus, F_o = " reference dust flux" at the height of one meter at a friction velocity of one meter per second.

and $$C_1 = \frac{F_o}{0.12\,\frac{m}{sec}}\left[\frac{u_*}{u_o}\right]^{\gamma} \tag{73}$$

$$F_1 = F_o\left[\frac{u_*}{u_o}\right]^{\gamma+1} \tag{74}$$

The above is known as the "Gillette and Shinn" model. The subscript "1" may be dropped in this expression if it is assumed that the vertical flux remains constant to within a few tens of meters from the ground.

With additional data provided by Gillette, tentative relationships were constructed between F_o, γ, and the soil conditions as expressed by I, "Soil Erodibility Index", a function of percentage of soil fractions greater than 840 micrometer in diameter (See Table 8). Figure 11 (from Shinn et al., 1974) shows these relationships. Their applicability can be gaged by means of a simple example.

It is easily seen from Figure 11 that F_o is a simple function of I, which the author interprets, roughly, as

$$F_o = \left(0.0276\,\frac{mg}{m^2 sec}\right) I \tag{75}$$

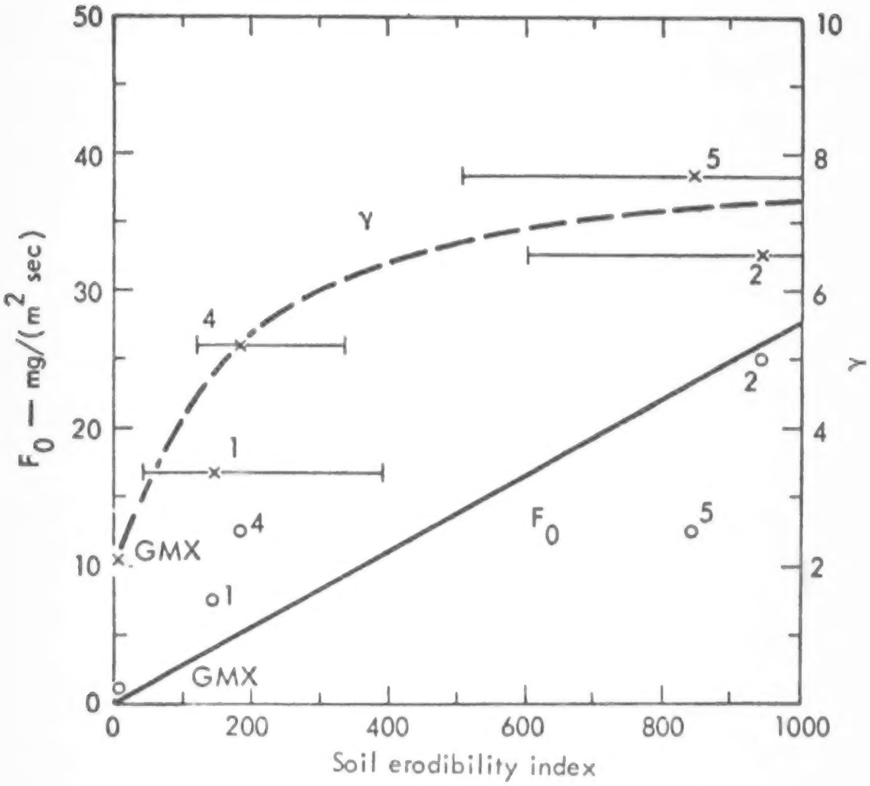


Figure 11. "Reference Dust Flux" F_o and exponent γ as functions of the soil erodibility index [From Shinn et al. (1974)]

To obtain $F_o = 0.73$ [GMX, Eq. (66)] the erodibility index in Eq. (75) would have to be $I = 26.45$, which, from the graph, corresponds to an exponent "γ" approaching the value 2.09 obtained empirically. However, with $\gamma = 6.38$ [Texas, Eq. (67)], the same graph would predict $F_o \simeq 12$, which is quite distant from the value 62.64 indicated for the Texas site.

With the intent of eliminating the inconvenience attendant to the use of graphs, the author of the present report has taken the liberty of reducing the tentative relationships indicated by these graphs to a set of equally tentative equations, as follows:

1. The relationship of γ to F_o as shown in Fig. 11 can be expressed empirically as

$$\gamma = \frac{-28 \text{ mg}/(\text{m}^2.\text{sec})}{4.1 \text{ mg}/(\text{m}^2.\text{sec}) + F_o} + 8.25 \qquad (76)$$

   The plot of $\gamma(F_o)$ as obtained from the above expression is presented in Figure 12, accompanied by the original relationship as redrawn from Fig. 11.

2. The correlation between F_o and I has already been shown to be, from Fig. 11,

$$F_o = 0.0276 \text{ mg}/(\text{m}^2\cdot\text{sec}) \times I \qquad (77)$$

3. Using equation (77), equation (76) can be rewritten as

$$\gamma = \frac{-28}{4.1 + 0.0276\, I} + 8.25 \qquad (78)$$

   The agreement of this expression with the original graph in Shinn et al. (1974) is shown in Figure 13, the dotted line indicating Shinn's original plot, the circles representing values obtained by equation (78).

4. Applying these relationships to equations (73) and (74), it follows that these can now be rewritten directly in terms of I as

$$C_1 = 0.23 \frac{\text{mg}}{\text{m}^3}\, I \left[\frac{u_*}{u_o}\right]^{\left(\frac{-28}{4.1 + 0.0276\, I} + 8.25\right)} \qquad (79)$$

$$F = 0.0276 \text{ mg}/(\text{m}^2\cdot\text{sec})\, I \left[\frac{u_*}{u_o}\right]^{\left(\frac{-28}{4.1 + 0.0276\, I} + 9.25\right)} \qquad (80)$$

   where I = erodibility index, dimensionless
   u_*= friction velocity, meters/second
   u_o= reference friction velocity = 1 m/sec

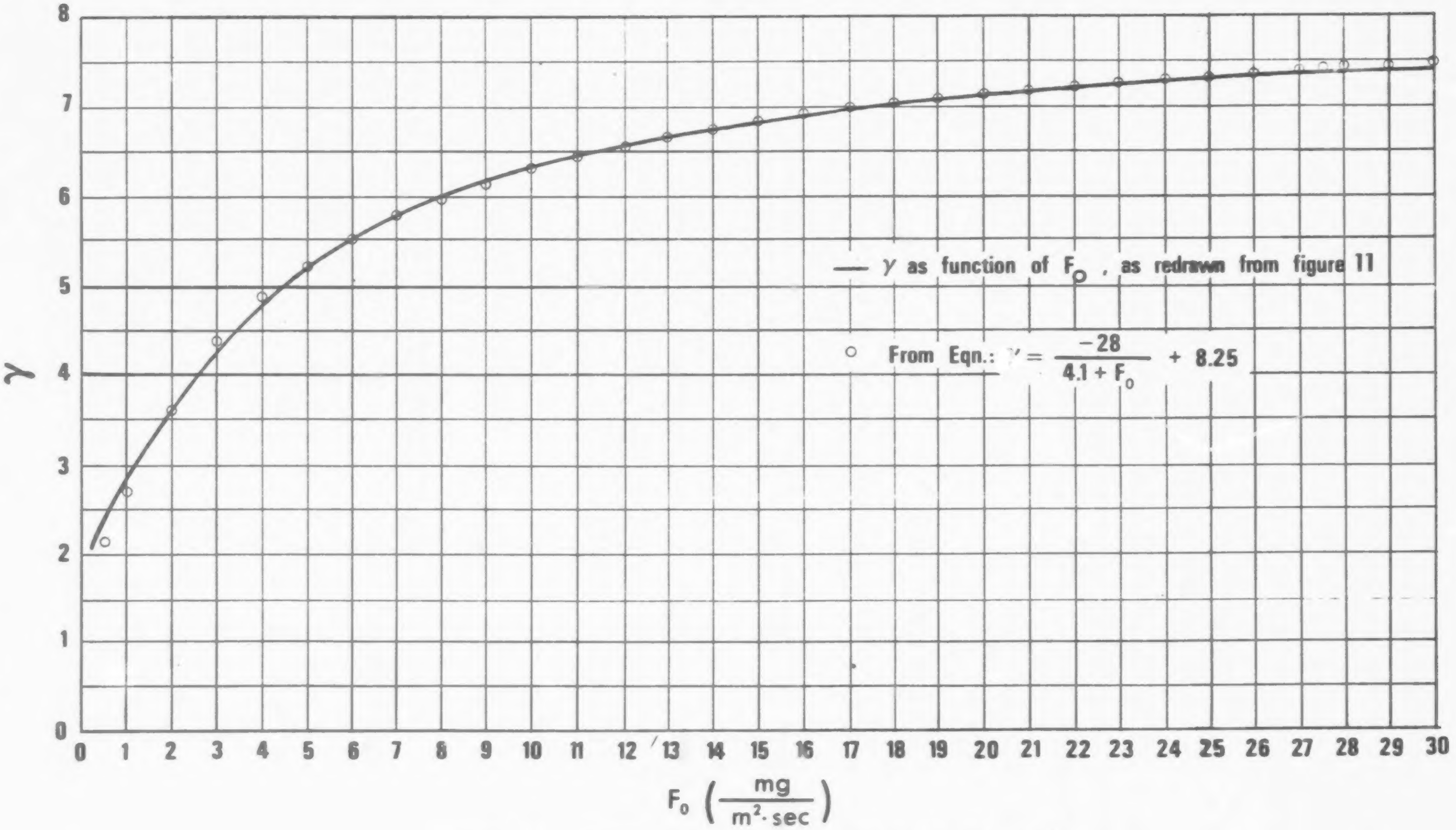
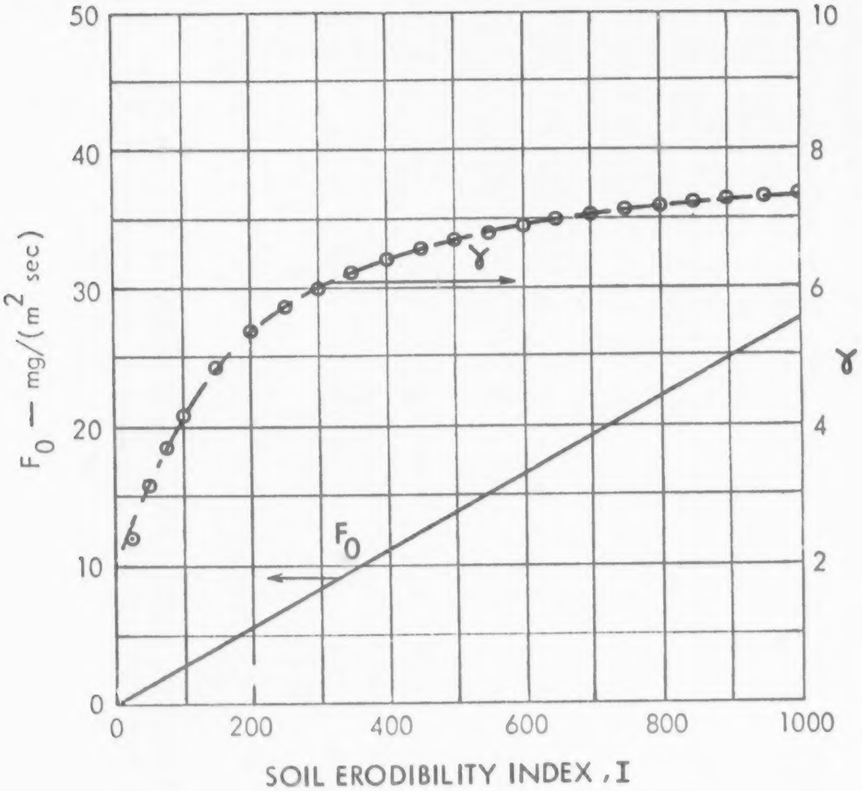


Figure 12. Exponent γ expressed as a function of F_o



○ From $\gamma = \dfrac{-28}{4.1 + 0.0276\,I} + 8.25$

— — From Fig. 11

Figure 13. Exponent γ of Gillette and Shinn model expressed as function of erodibility index I

TABLE 8. SOIL ERODIBILITY INDEX I BASED ON PERCENTAGE OF SOIL FRACTIONS GREATER THAN 840 MICROMETERS IN DIAMETER [Adapted from Chepil and Woodruff (1959)]

	Soil Erodibility Index, I									
0	70	80	100	120	150	175	280	450	1000	--
1	25	27	30	33	36	39	43	17	51	55
2	13	14	15	16	17	18	19	20	21	23
3	6.5	7.0	7.5	8.0	8.5	9.0	9.5	10.5	11.0	12.0
4	3.1	3.4	3.7	4.0	4.3	4.9	4.9	5.2	5.6	6.0
5	1.1	1.2	1.3	1.4	1.6	1.8	2.0	2.2	2.5	2.8
6	0.55	0.60	0.65	0.70	0.75	0.80	0.85	0.90	0.95	1.0
7	0.12	0.14	0.16	0.20	0.25	0.30	0.35	0.40	0.45	0.5
8	0.02	0.03	0.03	0.04	0.05	0.06	0.07	0.08	0.09	0.10
9										0.02
Tens / Units	9	8	7	6	5	4	3	2	1	0

Percentage of soil fractions greater than 840 micrometers in diameter

5. For completeness sake it should be mentioned that, on the basis of the above relationships, the empirical constant "a" of equation (62) can also be related to I directly by

$$a = 0.23\ I$$

The application of these equations should be limited to dry, sandy, unvegetated areas, similar to those at which the data was obtained. Coupled with a suitable model of atmospheric transport and diffusion, either equations (79) and (80) or equations (73) and (74) used in combination with the graph in Figure 11 could be applied to calculate airborne concentrations of plutonium within and without the contaminated area.

RESUSPENSION RATES

1. HEALY AND FUQUAY MODEL

One early model combining atmospheric transport and diffusion with contamination "pickup" rate was proposed by J. W. Healy and J. J. Fuquay (1959). The model assumes that the rate of pickup of particles from a surface is directly proportional to the ratio of wind forces to gravity forces on individual particles and, naturally, to the number of particles on the ground. Taking the wind force on a particle as proportional to the square of the wind velocity and to the particle area exposed to the wind, the rate at which particles become airborne, Q, is given by

$$Q = Sk' \frac{u^2 a}{\rho V} = SK' \frac{u^2 (\pi d^2)}{\rho (1/6 \pi d^3)} = SK \frac{u^2}{\rho d} \qquad (81)$$

where Q = rate of pickup, number of particles (#) per sec. (or g/sec)

a = area, $[m^2] = \pi d^2$, where d = diameter, m , of contaminant

V = volume, $[m^3] = \pi d^3/6$

ρ = density, g/m^3, of contaminant

S = number of particles on the ground, # (or grams)

K = "surface pickup coefficient," gms.sec/m^4

= 6K'

u = wind velocity, m/sec

Further, the model defines a "coefficient of deposition," Λ, reflecting the relationship of the rate of deposition on a ground surface due to turbulent diffusion, electrostatic effects and gravitational settling to the air concentration a short distance above the ground .

$$\Lambda = \frac{\text{Rate of deposition per unit area of ground per second}}{\text{Volumetric concentration per unit volume of air}} \quad (82)$$

where Λ = "coefficient of deposition," m/sec

Because deposition was defined to be dependent on the volumetric concentration very close to the ground, Sutton's expression for the concentration downwind from a continuous point source was used with z = 0, as follows:

specifying z = 0,

$$C(x,y,o) = \frac{RQ}{\pi C_S^2 u x^{2-n}} \exp\left(\frac{-y^2}{C_S^2 x^{2-n}}\right) \quad (83)$$

where C(x,y,o) = concentration in grams/m^3 at position x,y, meters

Q = rate of emission, grams/sec.

$C_S = C_Z = C_y$ = Sutton's virtual diffusion coefficient, $m^{n/2}$

n = stability coefficient, dimensionless

R = reflection factor to account for the presence of the ground

= 2 in this and following equations

u = wind velocity, m/sec.

The rate of depletion of the total quantity of material downstream due to fallout was determined using Λ as defined previously in Chamberlain's source depletion equation,

$$Q(x) = Q \exp\left(\frac{-4\Lambda x^{n/2}}{\sqrt{\pi}\, C_S u n}\right) \tag{84}$$

where Q = source rate, g/sec. at point source,

Q(x) = effective source rate, after depletion between point source and distance x from point source

Combining equations (81), (83), and (84) results in the following expressions:

1. Concentration downwind from a point source S_p in units of grams, particles, or μCi's

$$C(x,y,o) = \frac{2\,K\,S_p\,u}{\pi\, d C_S^2 \rho x^{2-n}} \exp\left(\frac{-4\Lambda x^{n/2}}{\sqrt{\pi}\, C_S u n}\right) \exp\left(-\frac{y^2}{C_S^2 x^{2-n}}\right) \tag{85}$$

Integrating (85) over y from $-\infty$ to $+\infty$ results in:

2. Concentration downwind from an infinite line source, with linear density of pollutant S_L in units of grams (or microcuries, or particles) per meter:

$$C(x,y,o) = \frac{2\,K\,S_L\,u}{\sqrt{\pi}\, d\rho C_S x^{(2-n)/2}} \exp\left(-\frac{4\,\Lambda\,x^{n/2}}{\sqrt{\pi}\, C_S u n}\right) \tag{86}$$

Integrating along the downwind coordinate up to a distance x_o produces:

3. Concentration downwind from a source infinite in the crosswind direction, but extending to a distance x_o downwind, with a surface concentration S_A in units of gms/m² or μCi/m²,

$$C(x,y,o) = \frac{K\,S_A u^2}{d\rho\Lambda}\left[\exp\left(-\frac{4\Lambda x^{n/2}}{\sqrt{\pi}\, C_S u n}\right) - \exp\left(-\frac{4\Lambda x_o^{n/2}}{\sqrt{\pi}\, C_S u n}\right)\right] \tag{87}$$

Some caution must be exercised in applying equation (87) to a physical case, in view of the somewhat misleading definition of x_o as "downwind extent of the source area." Presumably this indicates a relationship such as shown in Figure 14 below:

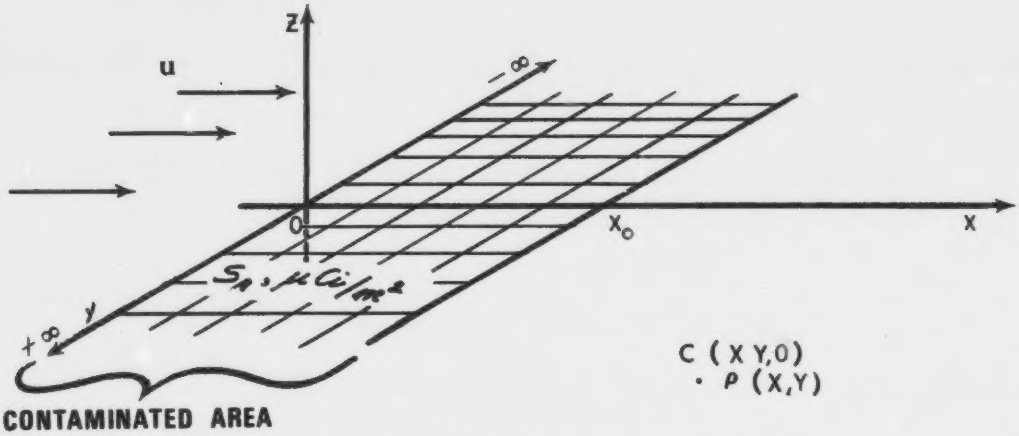


Figure 14. Presumed relationship of distance parameters in Healy and Fuquay equation of airborne concentration downwind from an area of finite downwind extent [Eqn.(87)]

$$\text{For } x \geq x_o,\ \exp\left(-\frac{4\Lambda x^{n/2}}{\sqrt{\pi}\, C_S un}\right) \leq \exp\left(-\frac{4\Lambda x_o^{\,n/2}}{\sqrt{\pi}\, C_S un}\right) \qquad (88)$$

This would mean that the bracketed term in equation (87) is either negative or zero, and the largest concentration C at point x,y,o is zero, when $x = x_o$, and negative in all other cases.

The true intent of equation (87) may be gaged by repeating the probable derivation of the equation with the following considerations:

a. The concentration at (x,y,o) due to an infinite line source at x' is:

$$C(x,y,0) = \frac{2\,K\,S_L\,u\,e^{-\left[\frac{4\,\Lambda\,(x-x')^{n/2}}{\sqrt{\pi}\,C_S\,u\,n}\right]}}{\sqrt{\pi}\,d\,\rho\,C_S\,(x-x')^{(2-n)/2}} \tag{89}$$

b. The differential concentration dC at (x,y,o) due to an increment dS_L at x' is:

$$dC(x,y,0) = \frac{2\,K\,u\,e^{-\left[\frac{4\,\Lambda\,(x-x')^{n/2}}{\sqrt{\pi}\,C_S\,u\,n}\right]}}{\sqrt{\pi}\,d\,\rho\,C_S\,(x-x')^{(2-n)/2}}\,dS_L \tag{90}$$

c. dS_L [μCi/m] can be interpreted as an areal density S_A [μCi/m^2] times an increment of distance dx': $dS_L = S_A\,dx'$

$$\text{and so, } dC = \frac{2\,K\,u\,S_A\,e^{-\left[\frac{4\,\Lambda\,(x-x')^{n/2}}{\sqrt{\pi}\,C_S\,u\,n}\right]}}{\sqrt{\pi}\,d\,\rho\,C_S\,(x-x')^{(2-n)/2}}\,dx' \tag{91}$$

d. To determine the total contribution of the area between x' = o and x' = x_o, to the concentration at (x,y,o), the above equation is integrated with respect to x' between x_o and o:

i) In $$C(x,y,0) = \int_0^{x_0} \frac{2\,K\,u\,S_A\,e^{-\left[\frac{4\,\Lambda\,(x-x')^{n/2}}{\sqrt{\pi}\,C_S\,u\,n}\right]}}{\sqrt{\pi}\,d\,\rho\,C_S\,(x-x')^{(2-n)/2}}\,dx'$$

ii) let $w = (x-x')^{n/2}$

and thus $dw = -\frac{n}{2}(x-x')^{(n/2-1)}\,dx'$

iii) then $$-\frac{2}{n}\,dw = \frac{dx'}{(x-x')^{(1-n/2)}} = \frac{dx'}{(x-x')^{(2-n)/2}}$$

iv) and equation i) becomes

$$C(x,y,0) = -\frac{4\,K\,S_A\,u}{\sqrt{\pi}\,d\,\rho\,C_S\,n}\int_{w=x^{n/2}}^{w=(x-x_0)^{n/2}} e^{-\frac{4\,\Lambda\,w}{\sqrt{\pi}\,C_S\,u\,n}}\,dw$$

v) Performing the integration produces

$$C(x,y,0) = \frac{-\,4\,K\,S_A\,u}{\sqrt{\pi}\,d\,\rho\,C_S\,n} \times \frac{1}{\left[\frac{-\,4\,\Lambda}{\sqrt{\pi}\,C_S\,u\,n}\right]}\left[e^{-\frac{4\,w\,\Lambda}{\sqrt{\pi}\,C_S\,u\,n}}\,\Bigg|_{w=\,x^{n/2}}^{w=(x-x_0)^{n/2}}\right]$$

vi) which reduces to eqn. (92)

$$C(x,y,0) = \frac{K\,S_A\,u^2}{d\,\rho\,\Lambda}\left[e^{-\frac{4\,\Lambda\,(x-x_0)^{n/2}}{\sqrt{\pi}\,C_S\,u\,n}} - e^{-\frac{4\,\Lambda\,x^{n/2}}{\sqrt{\pi}\,C_S\,u\,n}}\right] \qquad (92)$$

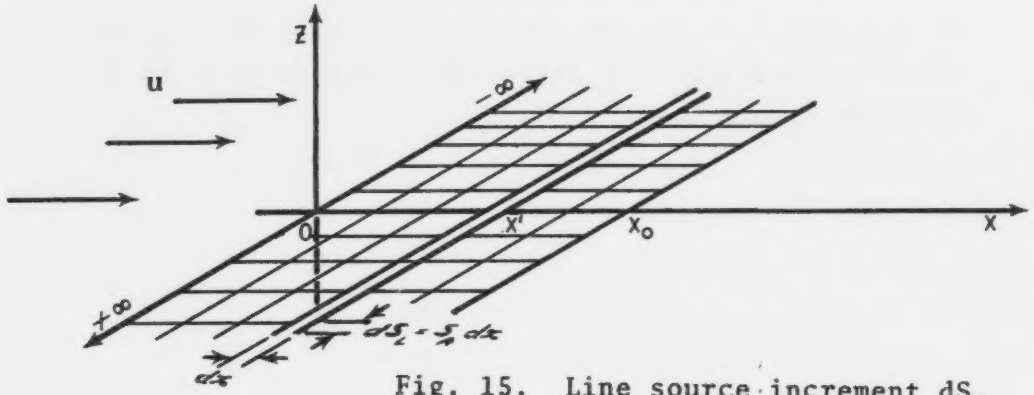


Fig. 15. Line source increment dS_L

The meaning of "x_o" as probably intended by Healy and Fuquay now emerges in true context from the above equation (92). If instead of "downwind extent of source area," x_o in equation (87) is interpreted as "distance between detector position x and downwind boundary of source area" or simply as "distance to the nearer boundary" ("$x-x_o$" in equation (91)), there is no conflict between equation (92) and equation (87).

Making use of equation (91) with the definitions:

$x_n = x-x_o$ = distance to nearer boundary of source area

$x_f = x$ = distance to farther boundary of source area,

$$C(x,y,0) = \frac{K\ S_A\ u^2}{d\ \rho\ \Lambda}\left[e^{-\left(\frac{4\ \Lambda\ x_n}{\sqrt{\pi} C_S un}\right)} - e^{-\left(\frac{4\ \Lambda\ x_f}{\sqrt{\pi} C_S un}\right)}\right] \qquad (93)$$

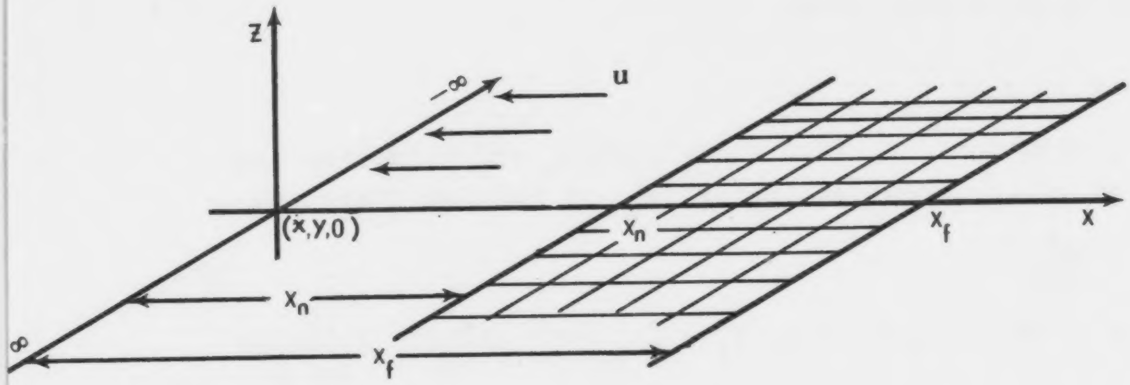


Figure 16. Revised interpretation of parameters employed in Healy and Fuquay equation for airborne concentrations downwind from an area of finite downwind extent [Eqn. (93)]

and taking into consideration that the concentration at x due to a source area extending from o to x_f-x_n is equivalent to the concentration at o due to a source extending from x_n to x_f, the concentration at o due to an infinite plane can be obtained.

Assuming that wind blows from only one direction at a given place and time, only half of the infinite plane in which the detector is located needs to be considered. Setting $x_n = o$ and $x_f = \infty$, the contribution from the semi-infinite plane extending from o to infinity produces

$$C = \frac{K\,S_A\,u^2}{d\,\rho\,\Lambda}\left(e^0 - \frac{1}{e^{\infty}}\right) \qquad (94)$$

or, 4. Concentration over an infinite area source

$$C(x,y,o) = \frac{K\,S_A\,u^2}{d\rho\Lambda} \qquad (95)$$

Note that the concentration C shows a dependence on the square of the wind velocity approximating that obtained experimentally by Sehmel at Rocky Flats, where

$$C = 0.45\,(u)^{2.1}$$

Rewriting equation (94) as $C = (Ku^2/d\rho\Lambda)S_A$ and recalling the definition of resuspension factor, R_f, it becomes clear that

$$R_f = \frac{Ku^2}{d\rho\Lambda} \qquad (96)$$

Recalling Slinn's expression, $R_f = \frac{3.5 \times 10^{-9}\ \text{cm/sec}}{1\ \text{ton/acre.month}}\left(\frac{r\dot{E}}{2\pi u\ S}\right)$

and ignoring the weak dependence of r on wind velocity,

$$R_f \propto \frac{\dot{E}}{u} \qquad (97)$$

From Bagnold and Chepil's soil erosion model,

$$q = C\rho u^{*3} \qquad (98)$$

where

q = rate of soil movement,gms/cm width sec
C = soil constant
u^* = drag velocity

If the assumption is made that E, rate of soil erosion, now in gms/cm^2.sec, is similarly proportional to u^{*3} and hence to u^3,

$$R_f \propto \frac{u^3}{u} \propto u^2 \qquad (99)$$

Thus, equation (95) would exhibit the same dependence on u as that implied in Slinn's model.

The relationship of u, wind velocity, to u^*, friction velocity, bears examination. Friction velocity may be defined by a modified form of the Prandtl equation as:

$$u = 5.75\, u^* \log_{10} \frac{Z}{k} + u_t \qquad (100)$$

where

Z = height at which u is measured, cm
k = "roughness" height, cm
u_t = fixed velocity at "roughness height," $\frac{\text{cm}}{\text{sec}}$
= impact threshold velocity

The roughness height is of the same order of magnitude as the height of a surface ripple formed by the sand flow. It is 0.3 cm for fine uniform sand, one cm for normal sand, 0.5 cm for clay and loam. u_t is the velocity at this height, fixed for a given diameter of the surface grains. u_t may be neglected only when the dust loading is very small and does not affect the velocity gradient with height (Stewart, 1967). Then the drag velocity u* is indeed proportional to u, and the relationships displayed in equation (81) through (95) are expected to hold.

That is, the resuspension should be strongly dependent on the wind speed only if $u_t \ll u$. This will occur if u itself is small and soil movement is slight. Stewart obtained a relationship of R_f to u^3 with wind velocities below 5m/sec and particles of size 0-8 micrometer and 0-12 micrometer. Healy and Fuquay obtained their values of K with wind velocities not exceeding 3.6 m/sec and particles of MMD = 7 μm.

The above discussion imposes some limitations on the applicability of Healy and Fuquay's model and by implication on Sehmel's, though not on Slinn's model, which did not explicitly relate erosion to wind velocity.

2. HEALY'S MODEL

The "surface pickup coefficient" K has units of gm.sec/m^4, a circumstance that may have influenced Healy's proposal of a different coefficient, called R_R for purposes of this discussion, such that:

$$R_R = \frac{K\ u^2}{d\rho} \qquad (101)$$

The units of the new coefficient are sec^{-1}; and from equation (81), restated below, the physical significance of the new parameter becomes clear:

$$Q = \frac{S\ K\ u^2}{\rho\ d}$$

$$Q = S\ R_R \qquad (102)$$

Since Q is the "rate of pickup," in μCi/sec or gms/sec and S is the total amount of contaminant present on the ground surface, in gms or μCi's, then R_R in sec^{-1} must be "that fraction of the contaminant deposit that resuspends per sec" or, simply put, a "resuspension rate."

Using equation (101) instead of equation (81) as a point of departure, and following essentially the same procedure outlined in equations (81) through (94), Healy's 1974 model can be obtained. This model is somewhat more general than Healy and Fuquay's 1959 version in the following aspects:

a. R_R is used instead of K. As already discussed, using K in conjunction with υ , ρ, d may conceivably restrict the applicability to velocities of less than 5 m/sec and low mass loading.

b. The effect of height Z above the ground in measuring concentrations is considered by including the corresponding Z-dependent terms in the diffusion equation.

c. C_Z is no longer assumed to be equal to C_y.

d. R is no longer explicitly restricted to a value of 2.

e. For completeness, the "coefficient of deposition" "Λ" is replaced by the more generally used term V_d "deposition velocity."

Applying these changes to equations (85), (86), and (87) results in expression equivalent to the ones following below:

1. Concentrations due to a point source containing S_p μCi's or grams of pollutant.

$$C = \frac{R\ R_R\ S_L}{C_y C_z \pi u\ x^{(2-n)}} \exp\left[\frac{-y^2}{C_y{}^2 x^{2-n}} + \frac{-z^2}{C_z{}^2 x^{2-n}}\right] \exp\left[\frac{-2RV_d x^{n/2}}{C_z u n \sqrt{\pi}}\right] \quad (103)$$

Air concentrations due to other source configurations may be obtained by integrating the above point source equation, by numerical techniques if the deposition pattern cannot be expressed as an equation.

2. Concentration due to an infinite line source upwind, with S_L μCi/m or gm/m of plutonium.

$$C = \frac{R\ R_R\ S_L}{\sqrt{\pi}\ C_z\ u^2\ x^{(2-n)/2}}\ e^{\left(\frac{-z^2}{C_z{}^2 x^{2-n}} - \frac{2\ R\ V_d\ x^{n/2}}{\sqrt{\pi}\ C_z\ u\ n}\right)} \qquad (104)$$

3. Concentration downwind from a source infinite in the crosswind direction, finite in downwind direction, with receptor at the ground level and x_1=distance to nearer boundary, x_2= distance to farther boundary of source area.

$$C = \frac{R_R\ S_A}{V_d}\left[e^{\left(\frac{-2\ R\ V_d\ x_1{}^{n/2}}{\sqrt{\pi}\ C_z\ u\ n}\right)} - e^{\left(\frac{-2\ R\ V_d\ x_2{}^{n/2}}{\sqrt{\pi}\ C_z\ u\ n}\right)}\right] \qquad (105)$$

Following the set pattern, the equivalent expression for an infinite area would be:

4. Concentration at any point near ground level from an infinite source area.

$$C = \frac{R_R\ S_A}{V_d} \qquad (106)$$

This permits establishing a relationship between resuspension factor and resuspension rate for a uniformly contaminated area.

$$C = \left(\frac{R_R}{V_d}\right) S_A = R_f\ S_A$$

$$R_f = \frac{R_R}{V_d} \qquad (107)$$

Furthermore, the expression $R_f\ S_A$, μCi/cm²-sec. or gm/cm²-sec., represents a flux of material, as pointed out by Anspaugh (1975). Using equation (61), this results in:

$$F = R_R S_S = -\ p\ k\ u_* C_1 \qquad (108)$$

$$\text{or} \quad R_R = -\ p\ k\ u_* \frac{C_1}{S_A} = -\ p\ k\ u_* R_f \qquad (109)$$

This obviously implies also that $V_d = p\ k\ u_*$ (110)

One additional equation was employed by Healy to describe concentration due to a Gaussian line source.

Concentration downwind from a line source with Gaussion distribution of material, with a standard deviation of A meters. The receptor is directly downwind from the peak ground concentration S_{L_p}

$$C = \frac{\sqrt{2}\ R\ A\ R_R\ S_{L_p}\ e^{\left(\frac{-z^2}{C_z{}^2 x^{2-n}} - \frac{2\ R\ V_d\ x^{n/2}}{\sqrt{\pi}\, C_z\ u\ n}\right)}}{\sqrt{\pi}\, C_z x^{(2-n)/2} u^2 \sqrt{C_y{}^2 x^{2-n} + 2A^2}} \qquad (111)$$

To utilize the above models, some judicious choice of values for the parameters R, n, C_Z, V_d, and R_R had to be made. These are given in Table 9, with exception of R_R which will be discussed later.

The Sutton parameters n, C_Z, C_y were chosen to correspond to unstable, neutral, and stable conditions. Healy made a comparison of the dispersion parameters σ_y and σ_Z as a function of distance using Pasquill's curves and the Sutton parameters used. These are shown in Figure 17 and Figure 18. Possibly no further comment is required on this account.

TABLE 9. VALUES OF V_d/u AS FUNCTION OF ATMOSPHERIC CONDITIONS, ROUGHNESS HEIGHT, REFLECTION FACTOR AND PARTICLE SIZE. [AFTER HEALY (1974)]

		ATMOSPHERIC CONDITIONS						
		UNSTABLE		NEUTRAL		STABLE		
SUTTON PARAMETERS	n	0.2		0.25		0.5		
	$C_y (cm^{n/2})$	0.45		0.2		0.3		
	$C_z (cm^{n/2})$	0.3		0.1		0.07		
SURFACE ROUGHNESS PARAMETER z_0(cm)	0.1	0.0093		0.0028		0.0046		FOR PARTICULATES OF SIZE > 1.5 μm
			0.0003		0.00008		0.00001	FOR PARTICULATES OF SIZE < 0.1 μm
	2.3		0.0005		0.00020		0.00009	
		0.017		0.0080		0.0029		
		1	1.97	1	1.97	1	1.97	
		REFLECTION FACTORS R=2−f (dimensionless)						

TABLE 9

$V_{d/u}$ VALUES AS FUNCTION OF n, C_y, C_z, z_0, R, Size

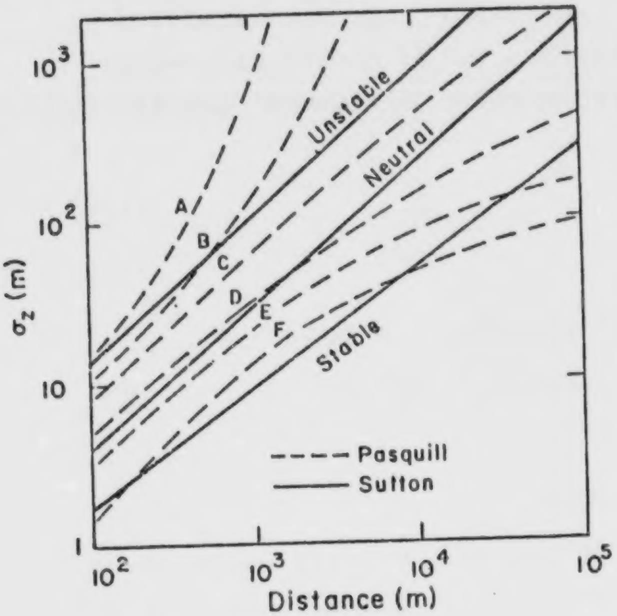


Fig. 17 Comparison of σ_z for Pasquill's curves and Sutton using parameters of Table 9

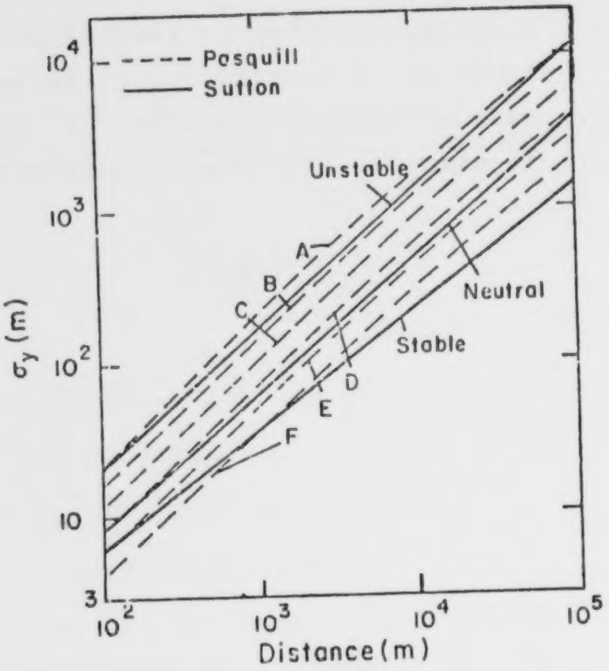


Fig. 18 Comparison of σ_y for Pasquill's curves and Sutton using parameters of Table 9

Adapted from "A Proposed Interim Standard for Plutonium in Soil," by J. W. Healy, LA-5483-MS, UC-41, (1974), pp. 35,36

The choice of values for the deposition velocity V_d and the related parameter R, reflection factor, was complicated by the general lack of readily usable data. Consequently, several assumptions were made by Healy that both limit the applicability of the model to a certain range of particles sizes and introduce possible inaccuracies within the applicable range.

Two mechanisms of settling are generally considered in deposition studies: that of gravitational settling, and that of turbulent transfer.

Gravitational settling describes the motion of a particle acted upon by gravity and aerodynamic drag, two opposing forces that eventually cancel each other with the result that a final limiting velocity is reached known as "terminal velocity," a function of particle size, shape, and mass. For the idealized case of homogeneous spherical particles up to 50-100 micrometer in diameter, the terminal velocity is given by Stokes' Law as:

$$V_g = \frac{2\ g\ r^2 \rho}{9 \kappa} \tag{112}$$

where

V_g = terminal (settling) velocity, cm/sec.

g = acceleration due to gravity, cm/sec^2

r = particle radius, cm

κ = air viscosity in poises.

For PuO_2 particles, of density 11.4 gms/cm^3, and 181 micropoises of air viscosity, the terminal velocities as function of size are given in Table 10.

TABLE 10. TERMINAL VELOCITIES OF PuO_2 SPHERICAL PARTICLES IN AIR

Radius (μm)	Terminal Velocity (m/sec)
0.05	.0000034
0.1	.000014
0.2	.000055
0.5	.00034
1.0	.0014
2.0	.0055
5.0	.034
10.0	.137
20.0	.548
50.0	3.43
100.0	13.72

For large particles, the terminal velocity overshadows any turbulence effects so that the deposition velocity is essentially equal to the terminal velocity. For smaller particles, the terminal velocity decreases to the point that turbulent eddies in the atmosphere exert sufficient force to overcome gravitational forces and retain the particle in suspension for long periods of time. The mechanism of deposition for these smaller particles is that of turbulent transfer, rather than that of gravitational settling.

Assuming the transfer of mass across the boundary layer of the atmosphere to be equal to the transfer of momentum, Fuquay (Healy, 1974) evaluated the transfer velocity to be:

$$V_t = \frac{(u^*)^2}{u_Z} \qquad (113)$$

where V_t = transfer velocity

u^* = friction velocity

u_Z = wind velocity at reference height Z.

The friction velocity u^* depends on the wind profile and the nature of the surface. For a neutral atmosphere, the relationship in equation (114) holds:

$$\frac{u_Z}{u^*} = \frac{1}{k} \ln \frac{Z}{Z_o} \qquad (114)$$

where k = Von Karman's constant

= 0.4

u_Z = wind velocity at reference height Z

Z_o = constant, characteristic of the surface. (height at which flow can be extrapolated to zero)

For stable or unstable conditions, Deacon (Healy, 1974) derived equation (115).

$$\frac{u_z}{u^*} = \frac{1}{k\,(1-\beta)}\left[\left(\frac{z}{z_o}\right)^{1-\beta} - 1\right] \qquad (115)$$

where $\beta > 1$ for an unstable atmosphere

$\beta < 1$ for a stable atmosphere

Assuming that Z_o is characteristic of the terrain and that it is the same for all stabilities as calculated for neutral conditions, and combining equation (113) with equations (114) and (115) results in:

for neutral conditions,
$$v_t = \left(\frac{k}{\ln \frac{z}{z_o}}\right)^2 u_z \tag{116}$$

for other conditions
$$v_t = \left[\frac{k\ (1-\beta)}{\left(\frac{z}{z_o}\right)^{(1-\beta)} - 1}\right]^2 u_z \tag{117}$$

Note that the assumption that Z_o is independent of stability and β is a function of stability does not necessarily imply that β is independent of Z_o. Healy, for simplicity, makes the assumption that β is independent of Z_o. Using values of β obtainable from the literature, Healy obtained a table of values, part of which is reproduced in Table 11.

TABLE 11. CALCULATED VALUES OF V_t/u

Z_o	Stable	Neutral	Unstable
0.1	0.00046	0.0028	0.0093
2.3	0.0029	0.0080	0.017

After J. W. Healy, in LA-5483-MS, UC-41, (1974) "A Proposed Interim Standard for Plutonium in Soils"

Comparing this table with Table 9 given previously, it is clear that these values correspond to V_d/u ratios for particulates of size >1.5 micrometer.

Assuming that particle diameter was the parameter used by Healy in determining size, it could be of interest to calculate the settling velocity obtainable from equation (112) for a PuO_2 spherical particle of size = 1.5 micrometer (radius = 0.75 micrometer).

$$V_g = \frac{2\ (980\ \frac{cm}{sec^2})\ (0.75 \times 10^{-4} cm)^2\ \ 11.4\ gms/cm^3}{9\ (0.000180\ gms/cm.sec)}$$

$$= 0.0776\ cm/sec$$

$$= 0.000776\ m/sec$$

Consequently, for particles of size greater than 1.5 micormeters,

$$V_g > 0.00078\ m/sec \qquad (118)$$

Since the "turbulent transfer velocity places an upper limit on the movement of the smaller particles through the boundary layer" (Healy, 1974), the values of V_t/u in Table 11 may be applicable provided that

$$(\frac{V_t}{u})\ u > V_g > 0.00078\ m/sec \qquad (119)$$

Using $V_t/u = 0.00046$ (lowest value in Table 11) in equation (119) results in

$$u > 1.7\ m/sec \qquad (120)$$

Thus, in a strict sense, use of Healy's model with the deposition velocity/average wind velocity ratios shown in Table 11 (and Table 9, for particles > 1.5 μm) requires placing a lower limit on the wind velocities for which the model is applicable.

Alternatively, an upper limit on the particle size to which the above ratios are applicable may be calculated as follows

$$\left(\frac{V_t}{u}\right) u > V_g$$

$$\left(\frac{V_t}{u}\right) u > \frac{2\ g\ r^2 \rho}{9\ \kappa} \tag{121}$$

$$\text{Size} = 2\ r < 3 \sqrt{\frac{2\ \kappa\ u}{g\ \rho} \left(\frac{V_t}{u}\right)} \tag{122}$$

$$\text{Size} < 54\ \mu\text{m} \sqrt{\frac{u}{1\ \text{m/sec}} \left(\frac{V_t}{u}\right)} \tag{123}$$

where g = 980 cm/sec^2

ρ = 11.4 gms/cm^3

κ = 180 x 10^{-6}poises (gms/cm.sec)

u = average wind velocity, in m/sec

(V_t/u) = transfer velocity/wind velocity ratio, from Table 11,

= dimensionless, function of Z_o and stability conditions

It follows that use of Healy's values of V_d/u for particles of size "greater than 1.5 micrometers" may not be appropriate under certain conditions. Their use is best left to the discretion of the user, via comparison of Tables 10 and 11 and equation (123), considering the margin of allowable error permissible in each specific application of the model.

Another parameter of importance, "retention efficiency," was considered in applying V_d/u's for small particles sizes. Assuming that particles of sizes " 1.5 micrometer" were relatively large, and that once deposited they would not "rebound" from the surface, Healy assigned to them a "retention efficiency" of

100 percent, and an overall V_d/u ratio equal to the V_t/u as shown in Table 9. Although the latter may be an underestimation of the actual deposition velocity for particles of this size, it could nevertheless provide a reasonable upper limit for smaller particles, with the proviso that retention efficiencies less than 100 percent be applied to these smaller particles, thus resulting effectively in smaller net deposition velocities.

From data available in the literature, Healy assumed that fission products generated by an electric arc in a given experiment had sizes of about 0.1 micrometer and a retention efficiency 3 percent, and that tracer particles used in another experiment (1 micrometer in size) had a retention efficiency of $f = 70$ percent. Excluding density from consideration due to lack of data, Healy further assumed a linear relationship of retention efficiency to particle size, as given in equation (124):

$$f = 0.74\ d - 0.04 \tag{124}$$

where d = particle size in μm.

The retention efficiency f was related to the reflection factor R by:

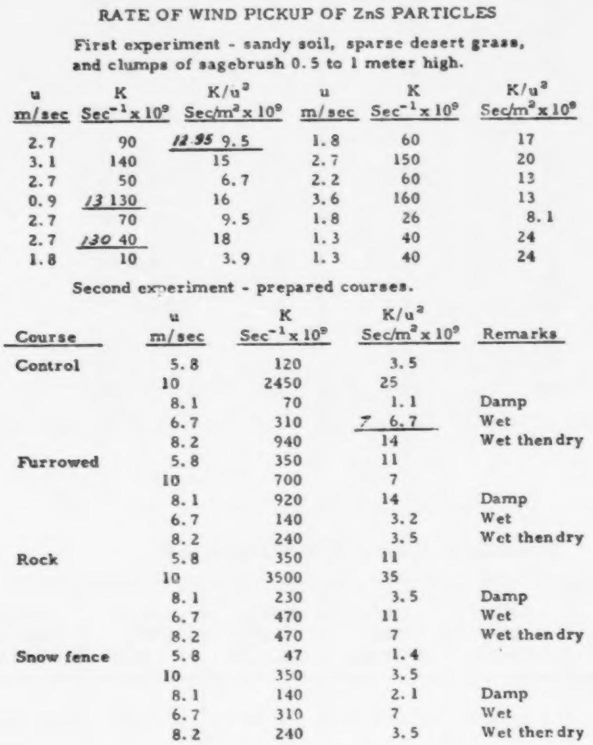
$$R = 2 - f \tag{125}$$

Thus, for f = 100 percent = 1, the reflection factor would be $R = 1$. For particles of size 0.1 micrometer, with $f = .03$, the reflection factor would be $R = 1.97$. These values are shown in Table 9, accompanied by deposition velocity - velocity ratios for ">1.5 micrometer" and "<0.1 micrometer" particle sizes. The latter were obtained, apparently, by multiplying the V_d/u ratios of the larger size particles by 0.03; this implies the further assumption that the retention efficiency does not decrease below three percent. Healy recognized that "new mechanisms such as electrostatic attraction, will come into play for the very small particles," although he used this fact to justify a relatively slow rate of decrease of the retention efficiency as a function of size, between 1.5 micrometer and 0.1 micrometer particles, rather than to postulate an increase in the retention efficiency for sizes less than some undetermined "critical size." Chepil (1945b) observed that the threshold velocity for initiation of soil movement is least for soil particles of about 0.1 millimeter to 0.15 millimeter in diameter, and increases for both larger and smaller particles. A similar increase in retention efficiency and corresponding V_d/u values for particles either larger or smaller than a given "critical size" does not appear to be altogether inconceivable. This and the preceding discussion of gravitational settling velocities versus turbulent transfer velocities for particles >1.5 micrometer lead the author to suggest a "range of best applicability" of Healy's model, using his values of V_d/u, such that 1.5 micrometer $\geq d \geq$ 0.15 micrometer where d = particle diameter, some caution being indicated for particles without this range.

Values of resuspension rate R_R were not provided explicitly by Healy for application in his model, although he reported some measurements made by other authors and values converted from the original "pickup coefficient" of the 1956 model (Healy & Fuquay, 1956) as order-of-magnitude estimates useful in further studies. The latter may be of particular interest, since some disagreement has been reported regarding the accuracy of Healy's values of R_R and R_R/u^2 (reported by Healy as "K" and "K/u^2") and which are shown in Table 12. Using the values of K (the present author's R_R) as basis, it is indeed apparent that the values of K/u^2 do not entirely correspond to the former, be it due to either computational or round-off error. This discrepancy is alleviated by referring to the original values of the "pickup coefficient", again K, (called K' by Healy in the 1974 work), as shown in Table 13, and relating the two (K' and K) by Healy's equation:

$$K = \frac{K'u^2}{\rho d} = 0.035\ k'\ u^2 \qquad (126)$$

TABLE 12. RESUSPENSION RATES (PICKUP COEFFICIENTS) USED IN HEALY'S MODEL (1974)



RATE OF WIND PICKUP OF ZnS PARTICLES

First experiment - sandy soil, sparse desert grass, and clumps of sagebrush 0.5 to 1 meter high.

u m/sec	K $Sec^{-1} \times 10^9$	K/u^2 $Sec/m^2 \times 10^9$	u m/sec	K $Sec^{-1} \times 10^9$	K/u^2 $Sec/m^2 \times 10^9$
2.7	90	12.95 9.5	1.8	60	17
3.1	140	15	2.7	150	20
2.7	50	6.7	2.2	60	13
0.9	13 130	16	3.6	160	13
2.7	70	9.5	1.8	26	8.1
2.7	130 40	18	1.3	40	24
1.8	10	3.9	1.3	40	24

Second experiment - prepared courses.

Course	u m/sec	K $Sec^{-1} \times 10^9$	K/u^2 $Sec/m^2 \times 10^9$	Remarks
Control	5.8	120	3.5	
	10	2450	25	
	8.1	70	1.1	Damp
	6.7	310	7 6.7	Wet
	8.2	940	14	Wet then dry
Furrowed	5.8	350	11	
	10	700	7	
	8.1	920	14	Damp
	6.7	140	3.2	Wet
	8.2	240	3.5	Wet then dry
Rock	5.8	350	11	
	10	3500	35	
	8.1	230	3.5	Damp
	6.7	470	11	Wet
	8.2	470	7	Wet then dry
Snow fence	5.8	47	1.4	
	10	350	3.5	
	8.1	140	2.1	Damp
	6.7	310	7	Wet
	8.2	240	3.5	Wet then dry

Reproduced from "A Proposed Interim Standard for Plutonium in Soils," by J. W. Healy. LA-5483-MS, UC-41 (1974), p.40, Los Alamos Scientific Laboratory

TABLE 13. SURFACE PICKUP COEFFICIENTS, COMPUTED USING HEALY AND FUQUAY MODEL (1959)

Estimate of K

Date	Distance (m)	Concentration (parts/m³)	Wind speed (m/sec)	*K*
29 Sept. 54	40	86	2·7	$3·7 \times 10^{-7}$
	40	120	3·1	$4·3 \times 10^{-7}$
30 Sept. 54	40	28	2·7	$1·9 \times 10^{-8}$
1 Oct. 54	40	14	0·9	$4·7 \times 10^{-7}$
5 Oct. 1954	61	9·2	2·7	$2·7 \times 10^{-7}$
	61	17	2·7	$5·1 \times 10^{-7}$
	40	4·6	1·8	$1·1 \times 10^{-7}$
	40	20	1·8	$4·9 \times 10^{-7}$
6 Oct. 54	61	16	2·7	$5·8 \times 10^{-7}$
	61	7·8	2·2	$3·6 \times 10^{-7}$
	61	1·4	3·6	$3·6 \times 10^{-7}$
8 Oct. 54	61	3·2	1·8	$2·3 \times 10^{-7}$
	61	6·4	1·3	$6·8 \times 10^{-7}$
	40	1·4	1·3	$6·8 \times 10^{-7}$

Estimates of K From Second Experiment

Period Surface condition Wind speed, 2 m	0–4 hr Dry 5·8 m/sec	4–5 hr Dry 10·0 m/sec	5–6 hr Damp 8·1 m/sec	6–8 hr Wet 6·7 m/sec	4–8 hr Dry–wet 8·2 m/sec
Control (grass)	1×10^{-7}	7×10^{-7}	3×10^{-8}	2×10^{-7}	4×10^{-7}
Furrow	3×10^{-7}	2×10^{-7}	4×10^{-7}	9×10^{-8}	1×10^{-7}
Rock	3×10^{-7}	1×10^{-6}	1×10^{-7}	3×10^{-7}	2×10^{-7}
Snow fence	4×10^{-8}	1×10^{-7}	6×10^{-8}	2×10^{-7}	1×10^{-7}
Average	2×10^{-7}	6×10^{-7}	6×10^{-8}	2×10^{-7}	2×10^{-7}

Reproduced from Health Physics Vol. 1, pp.433 and 434, (1959) by permission of the Health Physics Society.

Performing this computational exercise, it becomes readily obvious that the values of K and K/u^2 in Table 12 are both rounded-off results of performing the operation indicated by equation (126) on the values reported by Healy & Fuquay in 1956, shown in Table 13. The few exceptions noted by the present author could easily be due to clerical error. The author has taken the liberty of revising these by underlining the questionable values in Table 12 and providing the proposed value in parentheses. Possibly of more consequence is the fact that the values of K in the 1956 work (K' in the 1974 report by Healy) were obtained by assuming neutral atmospheric stability with $n = 1/4$ and $C_S = 0.18$, which correspond closely to the values employed by Healy in 1974 of $n = 0.25$, $C_y = 0.2$, and $C_Z = 0.1$ for the same atmospheric condition. Strictly speaking, however, this means that "pickup coefficients" calculated by Healy and Fuquay and later transformed into resuspension rates by Healy may not be applicable to other atmospheric conditions. Since the resuspension of pollutant is bound to be affected by the amount of turbulence near the ground surface, this restriction could be relaxed somewhat were it to be shown that atmospheric motions (such as characterized by average wind speed, for instance) had a minor effect on the magnitude of the resuspension rate. Clearly, this is not the case, as can be seen from Figure 19 , in which the resuspension rates obtained by Healy (as corrected by the author) and given in Table 12 are plotted against the average wind speed. Figure 19 indicates a definite trend towards greater values of R_R as the average wind velocity increases. For wind velocities of 3.6 m/sec and below, the relationship of the resuspension rate to wind velocity appears to be roughly parabolic, i.e., $R_R \propto u^2$. For velocities of 5.8 m/sec and above, the resuspension rate may be related to higher powers of the wind velocity, up to $R_R \propto u^{6+}$. It should be mentioned in this regard that Sehmel and Lloyd (1975) established a relationship of the resuspension rate with wind speed to the 6.5 power, for velocities above 3.6 m/sec and up to 20.1 m/sec. Referring to equations (102) through

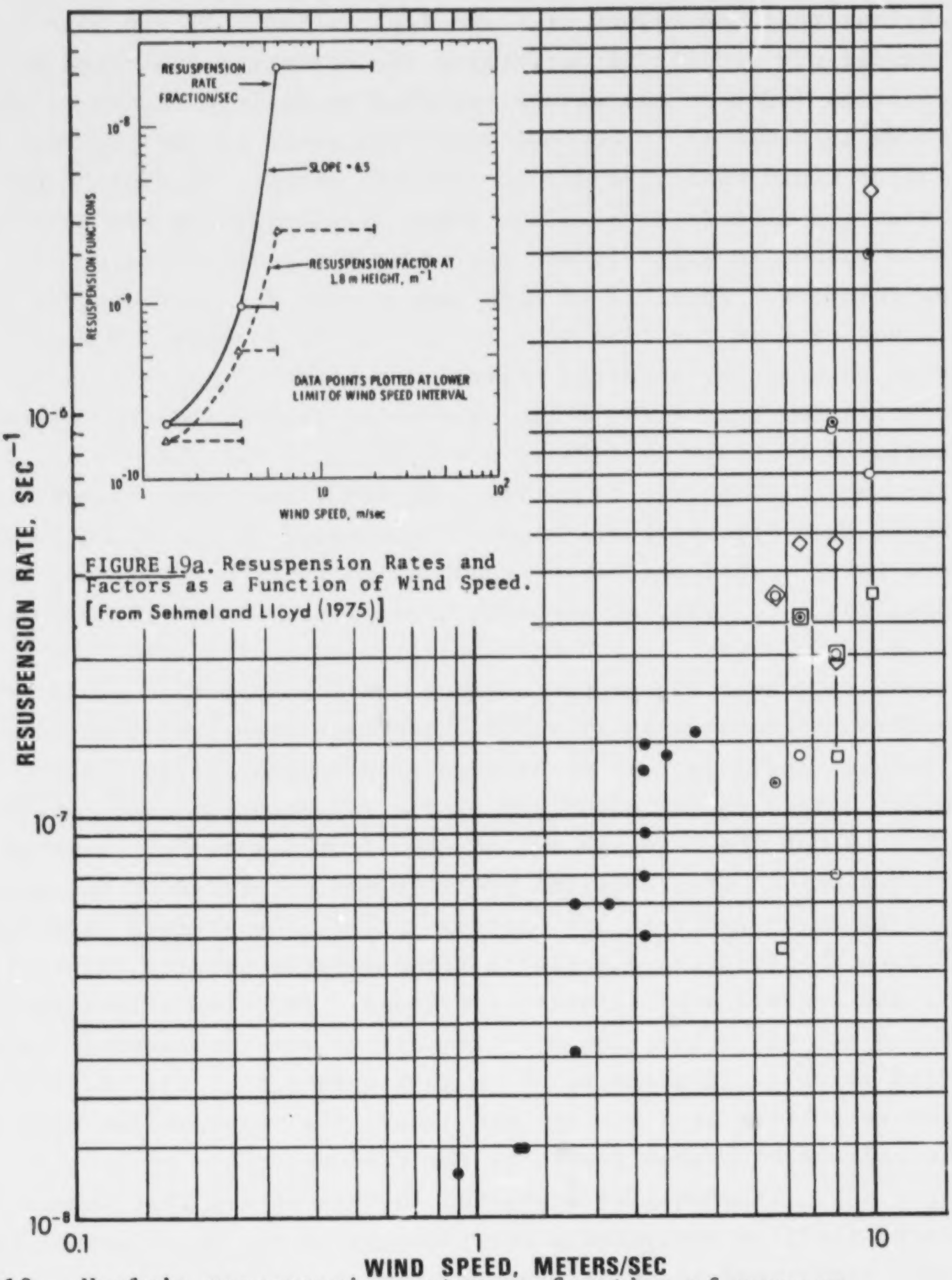


FIGURE 19a. Resuspension Rates and Factors as a Function of Wind Speed. [From Sehmel and Lloyd (1975)]

Figure 19. Healy's resuspension rates as function of average wind speed

(105), it is obvious that Healy's model does not include any such relationship of resuspension rate to wind speed. Consequently, each or any of the values of R_R (called K by Healy) in Table 12 can only be used in conjunction with the wind velocities and other conditions (assumed or observed) present in the derivation of these values. Attempts at greater generality using one specific value of R_R will result in errors in the air concentration of possibly several orders of magnitude.

Further examination of Healy's model reveals the presence of a term accounting for deposition of the resuspended material, as given by $\exp [-(2RV_d/\sqrt{\pi}\, C_z n\, u)x^{n/2}]$, but the absence of any expression explicitly indicating the further fortunes of the deposited material (re-resuspension). Presumably both the reflection factor R and the ratio V_d/u include consideration of this phenomenon through the dimensionless factor f known as the "retention efficiency" assumed to be 1.0 (100 percent) for particles equal or greater than 1.4 micrometer in diameter, and as low as .03 (3 percent) for particles equal or less than 0.1 micrometer in size. This would imply, apparently, that freshly "re-deposited" particles of greater than 1.4 micrometer in size cannot be "re-resuspended," whereas 97 percent of those particles equal or less than 0.1 micrometer in diameter are "re-resuspended" on impact, by literally bouncing off the surface. Since newly deposited (or "re-deposited") particles are supposedly easier to resuspend than those present on the ground for longer periods of time, the above reasoning would suggest that particles of $d > 1.4$ micrometer should seldom resuspend, or not at all, and that those found in suspension should range mostly from 1.4 micrometer to less than 0.1 micrometer. This would disagree with the size ranges reported by some investigators. Stewart (1967), for instance, mentions plutonium particles found in resuspension experiments to be 20-60 micrometer, and estimates < one percent in the hazardous-size range (≤3micrometer). Other results reported by the same author are not as conclusive. Following tests at

Maralinga, between 10 percent and 20 percent of the sampled activity was found on particles of less than micrometer size in diameter. Taking into account that a single particle of 50 micrometer diameter may be the source of as much activity as 125 million particles of 0.1 micrometer diameter, assuming homogeneous distribution, of the radionuclides, 20 percent of total activity may represent an enormous number density of small size particles. The only possible conclusion from the two quoted results is that no conclusion can be reached, at present, regarding the particle-size distribution of resuspended contaminant, and that any model that explicitly or implicitly assumes a distribution is subject to inevitable loss of generality.

It should also be noted that in his calculation of R_R, Healy used freshly seeded sources, which precluded the necessity of differentiating between a time-dependent surface concentration of pollutant $S_a(t)$ and a constant concentration per unit area S_A. Were the latter to be used, being constant with time and easier to measure, it would obviously have to be "weighed" with a factor accounting for weathering effects, a term such as exp $[-\lambda t]$, where λ and the reciprocal $T_{1/2}$ are the same parameters discussed in conjunction with the resuspension factor.

3. HORST'S MODEL

It will be recalled that resuspension rate was defined as the fraction of material lifted off the ground per unit time. This should be qualified, perhaps unnecessarily, by stating that it is the fraction of the material available for resuspension (by virtue of it not having become "fixed") and remaining from the resuspension process preceding a specific point in time. Thus two mechanisms are involved in reducing the concentration of material apt to be injected into the air stream, that of weathering and that of resuspension. Both these processes will proceed at rates proportional to the amount of remaining available material.

One additional mechanism needs to be considered in order to obtain the net rate of change of available concentration of contaminant - that of deposition. From the definition of deposition velocity V_d, the rate of increase of surface concentration can be seen to be equal to $V_d C(t)$. Thus the net rate of change of available surface concentration is:

$$\frac{dS_a(t)}{dt} = V_d C(t) - \lambda S_a(t) - R_R S_a(t) \qquad (127)$$

where

$S_a(t)$ = surface concentration available for resuspension, $\mu Ci/m^2$

$C(t)$ = air concentration, $\mu Ci/m^3$

V_d = deposition velocity, cm/sec

λ = fixation rate of available concentration, sec^{-1}

R_R = resuspension rate, sec^{-1}

This is precisely the approach used by Horst, Droppo, and Elderkin (1974)* in developing their resuspension model, with the qualification that the air concentration of interest C_r is that resultant from resuspension from an upwind source, and not the total air concentration used by Langham, Kathren, etc. In order to express $C_r(t)$ as a function of available surface concentration, $S_a(t)$, Horst et al. resorted to an expression similar to that in Langham's model, such that the two are related through a constant resuspension factor, R_F.

$$C_r(t) = R_F S_a(t) \qquad (128)$$

Introducing this relationship into equation (121) above, results in the expressions (129) and (130).

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* A similar expression, more general in form, was presented by A. J. Amato (Bibliographical reference-Amato, A.J...1971)

$$\frac{dS_a(t)}{dt} = (V_d R_F - \lambda - R_R)\ S_a(t) \tag{129}$$

$$S_a(t) = S_a(o)\ e^{(V_d R_F - \lambda - R_R)t} \tag{130}$$

where $S_a(o)$ = initial available surface concentration.

If the assumption were made that $C_r(t)$ is the only component of air concentration at a location where $S_a(t)$ is known, equation (130) would be identical to equation (10) discussed in connection with Langham's model.

$$S_a(t) = S_a(o)\ e^{-\lambda t} = S_a(o)\ e^{(V_d R_r - \lambda - R_R)t} \tag{131}$$

This would imply that $V_d R_F - R_R = 0$, or equivalently:

$$R_R = V_d R_F \tag{132}$$

Thusly, equation (122) could be rewritten as

$$C_r(t) = R_F S_a(o)\ e^{-\lambda t} \tag{133}$$

The difference between equation (133) and the expressions derived from Langham's and Kathren's models is academic, in that the latter do not specify the geographic origin of the resuspended contaminant; whereas Horst states that the source is "upwind." Indeed, in order to apply this "upwind" source to equation (129), the implicit assumption must be made that $S_a(t)$ upwind is equal to $S_a(t)$ at the point of interest, which makes the difference irrelevant.

Furthermore, if $S_a(t)$ is to vary uniformly, it is intuitively obvious that an infinite plane must be assumed. Otherwise, a depletion of the surface concentration at the upwind boundary of a seeded area would be coupled with an increase in the air concentration at the downwind edge with resultant gradients of $S_a(t)$ in the intervening areas.

Horst et al., however, were more cautious in their treatment of equation (129). They considered, in addition, the effects of deposition and resuspension on S_A, concentration per unit area ($S_A \neq S_a$).

$$\frac{dS_A(t)}{dt} = V_d C_r(t) - R_R S_a(t) \tag{134}$$

Soil fixation was excluded from the above equation, since it would not affect total concentration per unit area. Making use of equations (130) and (133), the above may be rewritten as follows:

$$\frac{dS_A(t)}{dt} = (V_d R_F - R_R)\, S_a(o)\, e^{(V_d R_F - \lambda - R_R)t} \tag{135}$$

$$S_A(t) = \frac{S_a(o)\ (V_d R_F - R_R)}{(V_d R_F - \lambda - R_R)}\, e^{(V_d R_F - \lambda - R_R)t} + C \tag{136}$$

where C is a constant of integration; at time t = o, the exponential term must equal 1, thus

$$C = \frac{S_A(o)(V_d R_F - \lambda - R_R) - S_a(o)(V_d R_F - R_R)}{V_d R_F - \lambda - R_R} \tag{137}$$

Recalling that $S_A(o) = S_a(o)$, the above provides:

$$S_A(t) = \frac{S_A(o)}{(V_d R_F - \lambda - R_R)} \left[-\lambda + (V_d R_F - R_R)\, e^{-(V_d R_F - \lambda - R_R)t} \right] \tag{138}$$

For $t \gg 1/(V_d R_F - \lambda - R_R)$, this reduces to equation (139).

$$S_A(t) \equiv \frac{S_A(o)\, \lambda}{\lambda - (R_R - V_d R_F)} \tag{139}$$

Since the "observed total concentration remains at better than 90 percent of the original,"

$$S_A(t) \gtrsim 0.9 S_A(o) \text{ for } t \ll 1/(V_d R_F - \lambda - R_R) \tag{140}$$

$$\frac{\lambda}{\lambda - (R_R - V_d R_F)} \geq 0.9 \tag{141}$$

$$\lambda \geq 0.9 \, [\lambda - (R_R - V_d R_F)] \tag{142}$$

$$R_R - V_d R_F \leq 0.1 \lambda \tag{143}$$

The above prompted Horst et al. to offer two possibilities:

ALTERNATIVE I

The difference $R_R - V_d R_F$ is much smaller than the "weathering" or "fixation" rate λ, though neither R_R nor $V_d R_F$ is necessarily much smaller than λ, individually.

$$R_R - V_d R_F \ll \lambda \tag{144}$$

This would lead to the already familiar conclusion that

$$R_R \equiv V_d R_F \tag{145}$$

ALTERNATIVE II

The magnitude of $R_R - V_d R_F$ is immaterial, given that both R_R and $V_d R_F$ are each independently much smaller than λ.

$$R_R \ll \lambda \tag{146}$$

$$V_d R_F \ll \lambda \tag{147}$$

By comparing a fixation rate of $2 \times 10^{-7}\,sec^{-1}$ (a 40-day half-life) to measured resuspension rates of 10^{-8} to 10^{-7} sec^{-1} and V_dR_F values ranging from 10^{-10} to 10^{-6} sec^{-1}, Horst et al. concluded that neither possibility could be rejected. The author feels that additional data could shed some light on the dilemma, although final resolution of it may have to be postponed until more conclusive evidence is available:

1. Healy and Fuquay (1959) calculated "pickup coefficients" of 10^{-8} - 10^{-7} gms-sec/cm^4, which translated into resuspension rates by means of equation (126) result in values ranging from 3.5×10^{-6} to 10^{-8} sec^{-1} (Healy, 1974).

2. Sehmel and Lloyd (1975) measured R_R's of 2×10^{-8} sec^{-1} to 2×10^{-10} sec^{-1} for "respirable" particles and one value of 1.3×10^{-11} sec^{-1} for "non-respirable" particles. The resultant range of values of the resuspension rate is consequently 3.5×10^{-6} sec^{-1} to 1.3×10^{-11} sec^{-1}. This corresponds to the range of V_dR_F values estimated by Horst et al.. However, the latter were calculated on the basis of initial resuspension factors 10^{-4} m^{-1} to 10^{-6} m^{-1}, and minimum deposition velocities 10^{-2} m/sec to 10^{-4} m/sec. Thus, higher V_d's are possible, which will serve to extend the upper limit of the V_dR_F range. In addition, values of R_F of 10^{-3} m^{-1} have been reported by Mishima (1964).

3. The value of λ used by Horst et al. was $2 \times 10^{-7}\,sec^{-1}$, equivalent to a 40-day half-time. However, λ is known to decrease with time. Anspaugh et al. (1973, 1975) observed half-times equivalent to 1×10^{-7} sec^{-1} and 3×10^{-8} sec^{-1}. Consequently λ should not be limited to one value, but rather to a range of values which from the above data is seen to be 2×10^{-7} sec^{-1} to 3×10^{-8} sec^{-1}. Since both V_dR_F and R_R may have values several

orders of magnitude greater than λ, Alternative II must be rejected on a general basis. Although there are instances in which $\lambda \gg R_R$ and $\lambda \gg V_d R_F$, these cannot be used to imply generality. By elimination, Alternative I should be correct. To be generally applicable, however, the relationship implied by Alternative I would have to hold under all conditions. The author knows of at least one instance, described in the following section, when this relationship is not maintained.

4. Sehmel and Lloyd (1975) made simultaneous measurements of resuspension rates and resuspension factors as functions of average wind velocity. For velocities greater than 3.6 m/sec, Sehmel and Lloyd calculated R_R to increase with wind speed to the 6.5 power (Figure 19a). From the graph, the present author estimated that R_F for the same interval varies as wind power to the 3.4 power. At lesser wind velocities, the dependences of R_R and R_F on wind speed appear to match more closely. However, the magnitude of R_F is less than that of R_R at all wind speeds, by factors of 1.1 to approximately 2 at low velocities, and up to an order of magnitude for the higher ranges shown in the graph. Consequently, in order for $R_R = V_d R_F$ exactly, the deposition velocities V_d would have to be greater than 1 m/sec at low wind velocities and as high as 10 m/sec at wind speeds exceeding 5.8 m/sec. These values are greatly in excess of deposition velocities generally reported in the literature.

The discussion in (4) suggests that Alternative I should be abandoned and, in fact, that Alternative II is a more reasonable assumption at low wind velocities. However, Alternative II was found questionable as consequence of the discussion in (3). That would indicate that neither possibility is generally applicable, which leaves the modeler with no possibility at all. In other words, it may not be generally true that $\lambda \gg R_R - V_d R_F$, and therefore the areal concentration S_A may change with time more rapidly than generally observed or expected.

Investigating the conditions under which $R_R \equiv V_d R_F$ should be expected to hold provides excellent reason for returning to Sehmel and Lloyd's observations, for which it does not. In their experiment, these investigators seeded an area of 22.9 m radius with calcium molybdate particles less than a micrometer in size. The resuspension factor was calculated via measurements made at a 1.8 meter height at the center of this small area. For larger areas, R_F should be higher, according to Sehmel and Lloyd, and the difference $R_R - V_d R_F$ correspondingly smaller, until, for an infinite area, no net loss of areal concentration S_A could be assumed.

$$\frac{dS_A}{dt} = (V_d R_F - R_R)\ S_a(o)\ e^{-[\lambda + R_R - V_d R_F]t} = 0 \qquad (148)$$

For the above condition to hold for any time t, there is no alternative but to postulate $V_d R_F = R_R$. Note that Horst et alii model implicitly assumes spatial uniformity of S_a, surface concentration of contaminant, a situation clearly obtainable with an infinite area, but of increasingly uncertain accuracy for progressively smaller areas. Stretching the argument, it can be said that Alternative I, $V_d R_F \equiv R_R$, should hold for "large" areas. The same should be true for the model of Horst et al.

Air concentration due to resuspension, according to this model, can be derived from $C(t) = R_F S_a(t)$ and equation (131).

$$C(t) = R_F S_a\ o)\ e^{-\lambda t} \qquad (149)$$

This is equivalent to Langham's and Kathren's expressions, as given by equations (9) and (11). Further, since $S_a(o) = S_A(o)$, it can be transformed to equation (150).

$$C_r(t) = S_A(o)\ R_F e^{-\lambda t} \tag{150}$$

Assuming that $S_A(t) \equiv S_A(o)$, as was done in eliminating $R_R - V_d R_F$ from the exponential term and the denominator in equation (139) produces equation (151).

$$C_r(t) \equiv S_A R_f(t) \tag{151}$$

$$\text{where } R_f(t) = R_F e^{-\lambda t} = R_f(o) e^{-\lambda t} \tag{152}$$

This corresponds to Anspaugh's model, with the qualification that the latter took into consideration λ being time dependent, whereas Horst's model does not. This suggests a further limitation on the applicability of the model presently discussed, that of validity for short time periods.

Exposure due to resuspension was determined by integrating equation (151) with respect to time.

$$E_r = \int_0^t C_r(t)dt = \frac{S_A(o) R_f(o)}{\lambda} (1 - e^{-\lambda t}) \tag{153}$$

Note that $S_A(o)$ originates from deposition from an original cloud of contaminant, passing over the area of interest in a time t,

$$S_A(o) = C_o T\ V_d \tag{154}$$

where

C_o = air concentration of original pollutant cloud, μCi/m^3

T = time of cloud passage, sec.

V_d = deposition velocity, m/sec.

The exposure to the original passage of pollutant is

$$E_o = C_o T \tag{155}$$

The ratio of exposure due to resuspension to that due to the original contaminant cloud would be

$$\frac{E_r}{E_o} = \frac{V_d R_f(o)}{\lambda} (1 - e^{-\lambda t}) \qquad (156)$$

By neglecting the exponential term after several half-lives, Horst et al. reduced equation (156) to

$$\frac{E_r}{E_o} = \frac{V_d R_f(o)}{\lambda} \qquad (157)$$

Note that this last simplification, that of $1-e^{-\lambda t} \equiv 1$, becomes increasingly valid with time, whereas $\lambda = \lambda(o)$ = constant in the denominator becomes increasingly invalid. This indicates the existence of an optimum time range for the application of equation (157), dependent on the actual values of the parameters involved and the time-dependence of λ.

Using 10^{-10} sec$^{-1} \leq V_d R_f(o) \leq 10^{-6}$sec^{-1} and $\lambda = 2 \times 10^{-7}$ sec, Horst et al. determined a range of values for the ratio:

$$5 \times 10^{-4} \leq \frac{E_r}{E_o} \equiv \frac{V_d R_f(o)}{\lambda} \leq 5 \qquad (158)$$

Using $R_f = 3 \times 10^{-4} m^{-1}$, reported by Mishima (1964) would result in an upper limit of 15, rather than 5.

Note that roughly the same results would be obtained by replacing $V_d R_F$ with R_R and using the R_R values calculated by Healy (1974) and Sehmel and Lloyd (1975) [see (1) and (2) in the preceding discussion].

$$4.8 \times 10^{-5} \leq \frac{E_r}{E_o} \equiv \frac{R_R}{\lambda} \leq 17.5 \qquad (159)$$

The situation is further complicated by Horst's use of $\lambda = 2 \times 10^{-7}$ sec. Lower values of λ are obtainable from Anspaugh's observations, such as 1×10^{-7} and 3×10^{-8} sec. The higher values of λ correspond to initial periods, within a few half-lives after deposition. Equation (159) includes a somewhat conservative upper limit for such periods. Use of the lower values of λ, strictly applicable to aged deposits, would result in an "extremely conservative" upper limit of the E_r/E_o ratio.

$$\frac{E_r}{E_o} \leq 116 \tag{160}$$

Horst et al. extended their model to include a situation in which "a continuous trace level release of material to the atmosphere produces a uniform air concentration" C_o. This required an alteration being made in equation (127).

$$\frac{dS_a(t)}{dt} = V_d C_r(t) + V_d C_o - R_R S_a(t) - \lambda S_a(t) \tag{161}$$

Noting that $V_d C_o$ represents a constant rate of increase in the available surface concentration $S_a(t)$, and that $V_d C_r(t) = V_d R_F S_a(t)$ transforms this equation to the following:

$$\frac{dS_a}{dt} + [\lambda + (R_R - V_d R_F)]\, S_a(t) = V_d C_o \tag{162}$$

This is a simple linear differential equation of the first order, solvable by means of the integrating factor $e^{[\lambda + R_R - V_d R_F)]\, t}$:

$$S_a(t) = e^{-[\lambda + (R_R - V_d R_F)]\, t} \left\{ C + \int V_d C_o \, e^{[\lambda + (R_R - V_d R_F)]\, t} \, dt \right\} \tag{163}$$

$$S_a(t) = \frac{V_d C_o}{[\lambda + (R_R - V_d R_F)]} + C\, e^{-[\lambda + R_R - V_d R_F)]\, t} \qquad (164)$$

Assuming that $S_a(o) = 0$, $C = - V_d Co/[\lambda + R_R - V_d R_F)]$

$$S_a(t) = \frac{V_d C_o}{[\lambda + (R_R - V_d R_F)]} \left\{ 1 - e^{-[\lambda + R_R - V_d R_F)]\, t} \right\} \qquad (165)$$

Applying once again the simplifying assumptions $\lambda >> R_R - V_d R_F$ and $e^{-\lambda t} \equiv o$ at $t >$ several half-lives,

$$S_a(t) \equiv \frac{V_d C_o}{\lambda} \quad \text{for } t > \text{several half-lives} \qquad (166)$$

Since $C_r(t) = R_F\, S_a(t)$, the concentration due to resuspension at any time t (more than several half-lives) is:

$$C_r(t) = \frac{R_F V_d C_o}{\lambda} \qquad (167)$$

$$\frac{C_r(t)}{C_o} = \frac{R_F V_d}{\lambda} = \frac{R_f(o) V_d}{\lambda} \qquad (168)$$

Naturally, this also implies that $E_r/E_o = R_f(o)V_d/\lambda$, as well, the same relationship described in equation (157).

RESUSPENSION RATIO (AMATO'S MODEL)

For the purposes of this report, equation (168) in the preceding section serves to introduce a new parameter, the resuspension ratio, defined as the ratio of air concentration due to resuspension to that arriving directly from some initial source, at a given location.

$$R_{r/d} = \frac{\text{C Resuspended}}{\text{C Direct}} \qquad (169)$$

In his 1975 paper, Amato dealt with a specialized case, that of a continuous source, after steady state had been established. His model treats the distribution and redistribution of pollutant along a single space coordinate, aligned with the horizontal wind vector, by considering a finite number of intervals on this coordinate. Within each interval i the concentration of pollutant is uniformly distributed both areally, on a surface of length L_i and infinite width, and volumetrically, down to a depth h (h ≅ top centimeter of topsoil), and varies with time only as a function of deposition, resuspension and downward migration into the soil. In the i-th interval, the rate of change of the contaminant concentration S_i is given by:

$$h\,\frac{dS_i(t)}{dt} = h \sum_{j=1}^{i-1} D_i^{\,j} S_j(t - T_i^{\,j}) - \frac{q^r S_i(t)}{\rho} - \frac{\lambda S_i(t)}{\rho} \qquad (170)$$

Where

S_i = interval i surface concentration, μCi/cm²

h = surface interval thickness

ρ = soil density, gms/cm³

λ = weathering coefficient, gms/cm².sec

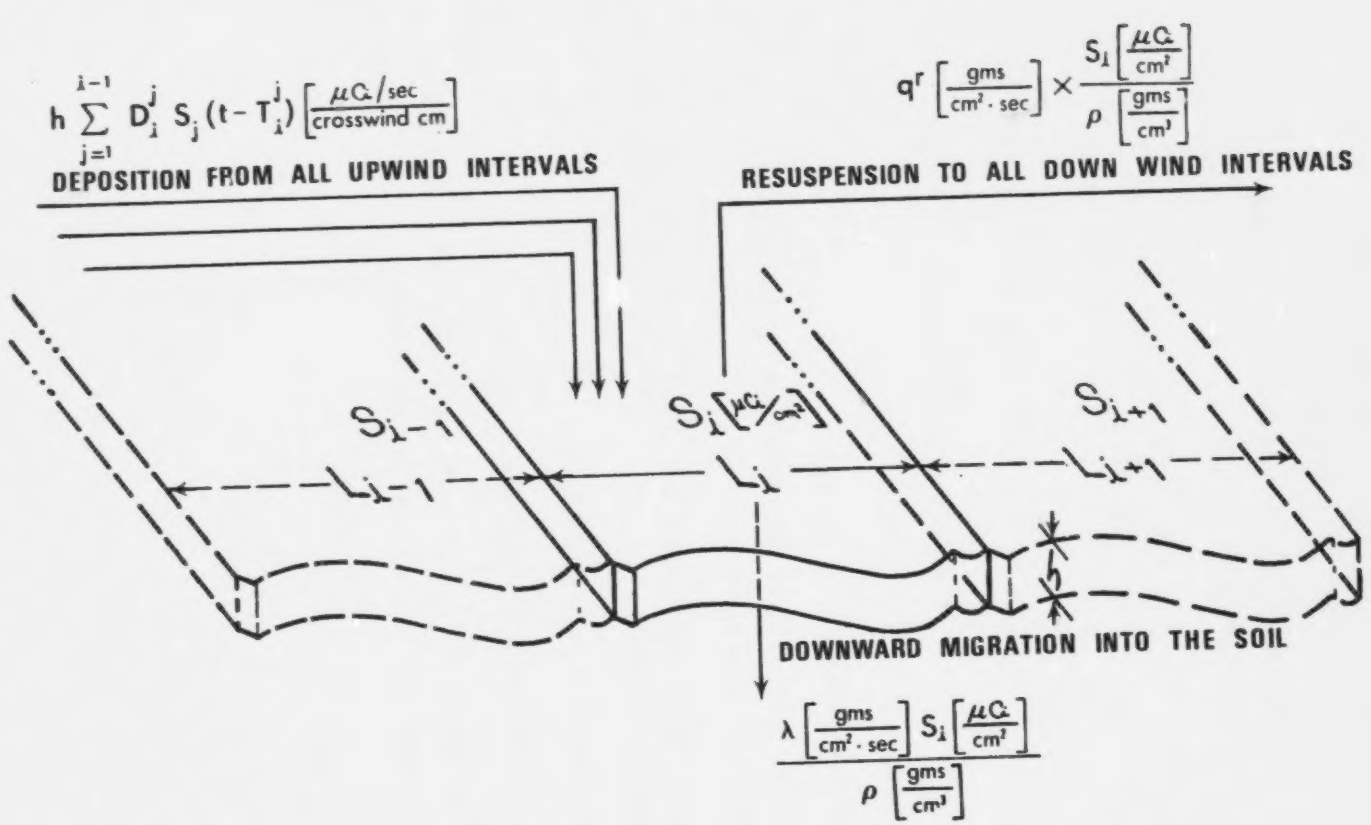

$h \sum_{j=1}^{i-1} D_i^j S_j (t - T_i^j) \left[\frac{\mu Ci/sec}{crosswind\ cm}\right]$
DEPOSITION FROM ALL UPWIND INTERVALS
$q^r \left[\frac{gms}{cm^2 \cdot sec}\right] \times \frac{S_i \left[\frac{\mu Ci}{cm^2}\right]}{\rho \left[\frac{gms}{cm^3}\right]}$
RESUSPENSION TO ALL DOWN WIND INTERVALS
S_{i-1}
$S_i [\mu Ci/cm^2]$
S_{i+1}
L_{i-1}
L_i
L_{i+1}
h
DOWNWARD MIGRATION INTO THE SOIL
$\frac{\lambda \left[\frac{gms}{cm^2 \cdot sec}\right] S_i \left[\frac{\mu Ci}{cm^2}\right]}{\rho \left[\frac{gms}{cm^3}\right]}$

Figure 20. Deposition, Resuspension and Downward Migration in a typical interval in Amato's model

q^r = soil flow rate for resuspension, gm/cm².sec

D_i^j = deposition coefficient of material on interval i due to resuspension from upwind internal j.

The deposition rate coefficient D_i^j is obtained by using a Sutton-Chamberlain diffusion model,

$$D_i^j = \frac{q^r}{\rho h} \left\{ e^{\left[\frac{-2 R V_d x_1^{n/2}}{\sqrt{\pi} C_Z u n}\right]} - e^{\left[\frac{-2 R V_d x_2^{n/2}}{\sqrt{\pi} C_Z u n}\right]} \right\} \quad (171)$$

Where q^r = q^s if j = 1, source soil flow rate for resuspension, gm/cm² sec.

R = 2-f = reflection factor, dimensionless

f = retention efficiency of soil

V_d = deposition velocity, cm/sec

x_1 = distance from center of interval i to nearer boundary of interval j.

x_2 = distance from center of interval i to further boundary of interval j.

C_Z = Sutton dispersion parameter

n = Sutton stability parameter.

The term T_i^j represents the time of flight particles resuspended from interval j must spend in traveling to interval i.

$$T_i^j = \sum_{k=j}^{i-1} \left[\frac{L_k + L_{k+1}}{2u} \right] \quad (172)$$

Where

L_k = length of interval k (=j,j+1, j+2...)

u = horizontal wind velocity (mean)

Only the first interval, representing a continuous ground source, is considered to be initially contaminated. For this interval, equation (170) is taken to be:

$$\frac{dS_1}{dt} = 0 \tag{173}$$

Consequently, the first term in the series of equation (170) becomes $q^s S_1 L_1/\rho h$, source release rate in µCi/sec/cm (crosswind). Note that q^s for the first interval is equivalent to q^r elsewhere, and has the same units.

The terms $\frac{"-q^r S_i(t)"}{\rho}$ and $\frac{"-\lambda S_i(t)"}{\rho}$ in equation (170) represent losses, due to resuspension and weathering, respectively. Solving the homogeneous equation

$$\frac{dS_i(t)}{dt} + S_i(t)\left[\frac{q^r+\lambda}{h\rho}\right] = 0 \tag{174}$$

will generate a solution representing the decrease of S_i due to resuspension and weathering.

The particular solution obtained by adding the incoming flow represented by $\sum_{j=1}^{i-1} D_i^j S_j(t-T_i^j)$ will then represent the increase in S_i due to transport from upwind intervals.

$$\frac{dS_i(t)}{dt} + S_i(t)\left[\frac{q^r+\lambda}{h\rho}\right] = \sum_{j=1}^{i-1} D_i^j S_j(t-T_i^j) \tag{175}$$

The complete solution $S_i(t)$ is obviously obtained by summing the complementary and particular solutions for each interval i. Foregoing further description of the method of solution, which would be, at best, a tedious repetition of the process described by Amato (1975), let it suffice to state that for long time intervals (i.e., approaching steady state conditions), the solution for a particular interval j becomes:

$$S_j(\infty) = S_1 \sum_{k=1}^{j-1} \frac{1}{(q^r + \lambda)^k} A_j^{j-k} \qquad (176)$$

Where

S_1 = constant, source concentration
A_j^{j-k} = constant, from particular solutions
$k = 1, 2, 3, \ldots i-1$

Treating each one of these intervals as an infinite crosswind source, the air concentration of soil reaching the i-th interval, $C_{s_i}^j$ is obtained by again using the diffusion model:

$$C_{s_i}^j = \frac{2q^r}{\sqrt{\pi}\, C_Z u} \int_{x_1}^{x_2} \frac{1}{x^{(2-n)/2}}\, e^{\left[\frac{-4\, V_d}{\sqrt{\pi}\, C_Z u} \frac{x^{n/2}}{n}\right]}\, e^{\left[-\frac{z^2}{C_Z^2 x^{2-n}}\right]} dx \qquad (177)$$

Note that the units of S_j are μCi/cm², and that dividing by h [cm] and ρ[gms/cm³] results in a term $S_j/h\rho$ [μCi/gm of soil]. Multiplying this expression by $C_{s_i}^j$ [gms of soil/cm³] results in a concentration of pollutant at interval i due to resuspension at j. Adding all the intervals upwind of i results in

$$C_{r_i} = \sum_{j=2}^{i-1} \frac{C_{s_i}^j S_j(\infty)}{h\rho} \qquad i = 3, 4, 5, \ldots. \qquad (178)$$

$$C_{d_i} = \frac{C_{s_i}^1 S_1}{h\rho} \qquad i = 2, 3, 4, 5, \ldots \qquad (179)$$

where the subscripts r, d, indicate "due to resuspension," and "directly from source," respectively.

Thus, the resuspension ratio for interval i may be written as

$$R_{r/d_i} = \frac{C_{r_i}}{C_{d_i}} = \frac{\dfrac{\sum_{j=2}^{i-1} C_{s_i}^{j} S_j(\infty)}{h\rho}}{\dfrac{C_{s_i}^{1} S_1}{h}} \qquad i = 3,4,5 \qquad (180)$$

where for i = 2, $R_{r/d_2} = 1$

Amato points out that the resuspension ratio is independent of both q^s and S_1 of the source release rate $q^s S_1 S_1/\rho h$, and further, if the weathering is negligible (if $\lambda << q^r$), then also independent of q^r, soil flow rate for resuspension, at steady state conditions.

Amato's model considered that redistribution of material which would occur through resuspension, and consequently saltation and creep were ignored. However, more than 90 percent of soil movement is assumed to occur precisely through these mechanisms, which suggests that the soil concentrations S_i may be severely underestimated, as should be seen from the following discussion.

Note that dividing equation (170) by h results in a resuspension term $(q^r/\rho h)\ S_i(t)$, and that q^r is the soil flow rate for resuspension, in units of gms/cm^2 . sec, which results in $(q^r/\rho h)$ having the same units as a resuspension rate, sec^{-1}. Specifically, since soil and not contaminant is being considered, it is obviously a "soil suspension rate" (i.e., that fraction of the top soil, to a depth h, which is suspended per unit time). Applying this rate to the contaminant with the evident expectation that, proportionately, an equal fraction of the total areal contamination should be suspended, results in $(q^r/\rho h)S_i(t)$. However, if the concentration S_i consists of resuspendible particles, a much greater fraction of the contaminant than of the soil itself should be resuspended.

On the other hand, if S_i were to include particles of all sizes, paralleling to some extent the particle-size distribution of the topsoil itself, the use of the term $(q^r/\rho h)S_i(t)$ would be justifiable, but that would necessitate the inclusion of saltation and creep effects in calculating S_i.

The only interval to which the expression $(q^r/\rho h)S_i(t)$ is strictly applicable is the first interval, assuming equal particle-size distributions of topsoil and contaminant or intimate mixing of the two. Amato used a different parameter, q^s, to represent the source flow rate, also in units of gms/cm²•sec. Since presumably the "soil suspension rate" (the present author's "$q^r/\rho h$") should be the same for both the source interval and for downwind intervals, the presence and differentiation between the quantities q^s and q^r leaves the author to suspect that $q^r/\rho h$ is not a "true" "soil suspension rate" but rather a parameter more akin to "resuspension rate of contaminant" R_R, discussed previously, to account for the higher susceptibility of deposited particles of resuspendible size to further resuspension.

Integrating equation (177) with the proviso that z = 0 results in

$$C_{S_i}^{j} = \frac{q^r}{V_d}\left[e^{\left(\frac{-2V_d}{\sqrt{\pi}C_z n u} x_1^{n/2}\right)} - e^{\left(\frac{-2V_d}{\sqrt{\pi}C_z n u} x_2^{n/2}\right)} \right] \qquad (181)$$

The units of $C_{s_1}^{j}$, suspended soil concentrations, are gms/cm³. Resuspended pollutant concentration over interval i can then be obtained by using equation (178).

$$C^{j}_{r_i} = C^{j}_{S_i}\ S_j(\infty)/\rho h$$

$$C^{j}_{r_i} = \frac{q^r S_j(\infty)}{\rho h V_d}\left[e^{\left(\frac{-2V_d}{\sqrt{\pi}C_Z nu} x_1^{n/2}\right)} - e^{\left(\frac{-2V_d}{\sqrt{\pi}C_Z nu} x_2^{n/2}\right)} \right] \tag{182}$$

Comparing this equation to equation (105) in the discussion of Healy's model, it becomes clear that, indeed

$$\frac{q^r}{\rho h} = R_R \tag{183}$$

The same expression would be obtained if q^r were replaced with q^s in solving the integral (177).

$$\frac{q^s}{\rho h} = R_R \tag{184}$$

The difference between q^r and q^s, not readily apparent from equations (183) and (184) above, is in their application to relatively fresh deposited contaminant, S_j, and an aged deposit in the source interval S_1, respectively. Note that the availability of resuspendible material has been modeled by Langham to be $S_a(t) = S_a(0)\ e^{-\lambda t}$, where S_a = surface contamination per unit area: it follows that the resuspension rate must be

$$\frac{q^s}{\rho h} = \frac{q^r}{\rho h}\ e^{-\lambda t} \tag{185}$$

where λ may be time dependent, $\lambda(t)$, in a fashion such as described in previous sections, although the author must admit to this being pure speculation on his part, since Amato (1974) does not mention these presumed relationships ($R_R = q^r/\rho h$ and $q^r \geq q^s$ or $q^s = q^r e^{-\lambda t}$).

TRAVIS'S MODEL

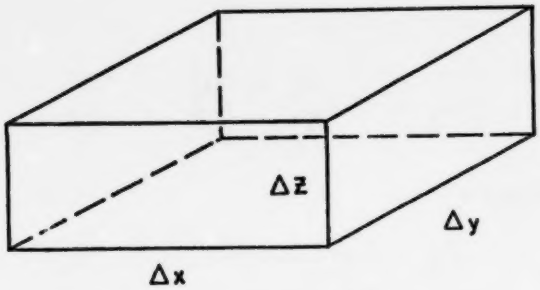
J. R. Travis (1975) provided a more comprehensive approach to resuspension modeling by considering the redistribution of contaminant two-dimensionally along the soil surface and including (implicitly) the effects of saltation and creep. The latter was done by assuming that the movement of mobile pollutants in particulate form is indistinguishable from that of the eroding soil in which they are distributed, which allowed Travis to apply the soil-erosion formulas developed by Bagnold, Chepil, Gillette, and others to the present problem.

By dividing the area of interest into a two-dimensional grid system, the movement of material from/into each of the cells comprising the planar mesh was seen to consist essentially of two components: one proceeding directly, characterized by a horizontal soil flux F_h of nonsuspendible material from/into an adjacent cell and one arriving/departing indirectly, by applying atmospheric dispersion and deposition models to material of suspendible size, lost by upwind cells to downwind cells at a rate expressed by a vertical flux F_v.

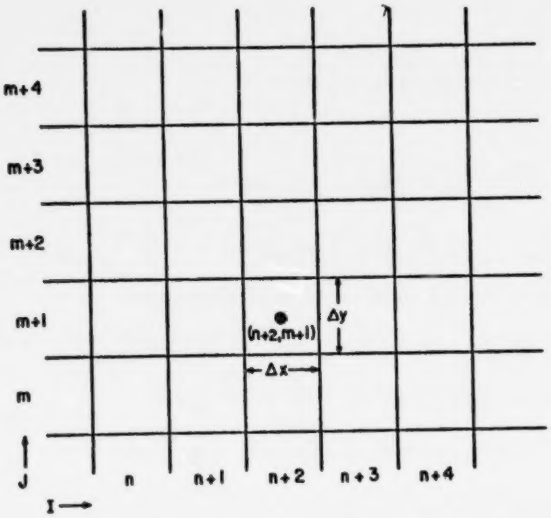
A control volume constructed atop a grid cell (Figure 21) permits application of mass conservation equations to the rate of change of material in the cell. Note, however, that air concentrations are not included in the material of concern - only that amount deposited or removed from the ground is considered.

Two alternative formulations were offered by Travis to describe the horizontal flux F_h:

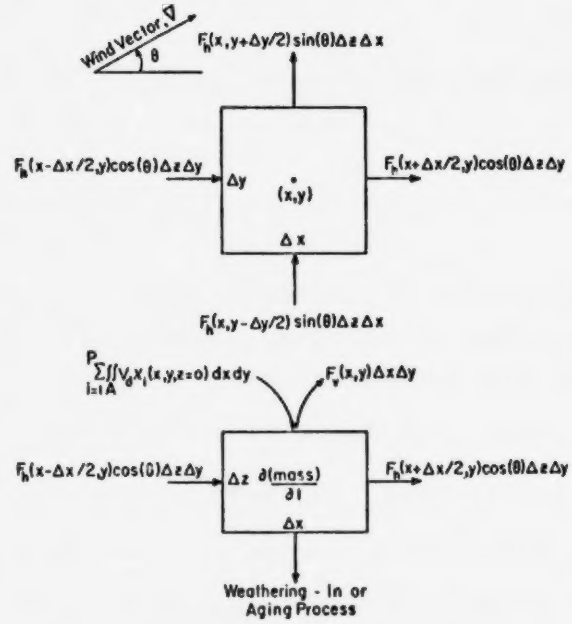
$$F_h = X \left(\frac{u_e^*}{u_r^*}\right)^3 \qquad (186)$$



Example of a control volume located on each mesh cell.



Two-dimensional mesh cell structure.



Control volume top and side view showing the contaminant mass flow.

The essence of this model is to describe the relocation of wind eroding soil-contaminant mixtures. This is accomplished by first characterizing soil and surface conditions within a square mesh structure overlayed on the surface. Then, according to certain meteorological conditions, a horizontal flux component transports material into adjacent mass conserving cells, and a vertical flux component generates puffs of suspendible particles. These puffs are emitted at regular time intervals. They diffuse downwind under time-dependent wind velocity and atmospheric stability conditions, maintaining during the time interval a three-dimensional Gaussian distribution of concentration with the cloud volume. The center of each Gaussian Puff configuration is determined by the trajectory since release, and its distribution is determined by the three dispersion coefficients σ_x, σ_y, and σ_z, which are increasing functions of travel distance. During the time interval, each cloud depletes material according to the area integral of the product of the deposition velocity and the average ground level concentration. Thus each puff has a center, volume, and material mass which are determined by the wind velocity, atmospheric stability, travel distance, and deposition velocity. After material is deposited on the soil, the contaminant redistribution processes, depending upon local conditions, may recycle to provide additional contaminant movement from these new sources.

Figure 21. Vertical and horizontal contaminant mass flows in/out of a typical cell in Travis' model [Adapted from Travis (1975)]

$$F_h = X \left(\frac{u_e^*}{u_r^*}\right)^2 \left(\frac{u_e^*}{u_r^*} - \frac{u_t^*}{u_r^*}\right) \qquad (187)$$

Where

F_h = horizontal soil flux, tons per rod width per hour

u_e^* = effective surface friction velocity, cm/sec.

$= \sqrt{\frac{\tau - \alpha}{\rho_a}}$, where τ = observed surface shear

α = surface soil moisture content

ρ = density of surface air, gm/cm

u_r^* = reference surface friction velocity, cm per sec..

$= \sqrt{\frac{\tau r}{\rho_a}}$, where τ = reference surface shear stress

= 33.1 dynes/cm^2 (Gilette et al. 1972)

u_t^* = threshold surface friction velocity, cm/sec, below which soil will not move.

X = soil erodibility friction, tons per rod per hour

$$= a \frac{I}{(RK)^b}$$

Where

a,b = constants, dimensionless, depending on past erosional history, type of residue, roughness, condition of the surface crust.

I = soil erodibility index (see Table 8)

R = amount of vegetative residue, lbs/acre

K = surface roughness equivalent, inches

The second of the equations given above provides for F_h approaching zero as the friction velocity decreases to $u_e^* \simeq u_t^*$. For $u_e^* < u_t^*$, F_h is taken to be zero.

The mass fraction of the contaminant in the horizontal flux was defined by Travis to be

$$fr_h = \frac{m_S + m_{NS}}{\rho\ h\Delta x\Delta y + m_S + m_{NS}} \quad (188)$$

Where fr_h = mass fraction of contaminant, dimensionless.

m_S = mass of suspendible contaminant in surface layer of depth h, gms.

m_{NS} = mass on non-suspendible contaminant in surface layer of depth h, gms.

ρ = density of native soil, gms/cm^3

$\Delta x\Delta y$ = area of grid cell, cm^2

h = depth of erodible surface layer, cm.

To account for the weathering process, Travis suggested two methods, one of which decreased effectively the amount of contaminant by means of a "weathering-in" function such as described by Anspaugh, et al. (1973). To follow this suggestion, it may be of some use to consider first the total mass of contaminant, m_S + m_{NS}, which can be related to total concentration per unit area, S_A, by

$$m_S + m_{NS} = \frac{S_A\Delta x\Delta y}{a_{SP}} \quad (189)$$

Where S_A = total concentration per unit area, μCi/cm^2 (assumed to be near the surface, top few cm)

$\Delta x\Delta y$ = area of grid cell, cm^2

a_{SP} = specific activity, μCi/gm.

Note, however, that all the parameters mentioned above are independent of time. Referring to the source mentioned by Travis, it can be recalled that the "weathering-in" function mentioned in this regard was that described by a half-time of 38 days, pertaining to the material decrease in availability to the resuspension process, not to erosion by saltation and creep. Furthermore, to apply this function to equation (188), the surface concentration $S_a(t)$ of pollutant would have to be considered, not the total concentration per unit area, S_A, which would signify that the depth h, to be realistic, would have to be extremely small, which may lead to unrealistically high values of fr_h, approaching $fr_h \simeq 1$ in the extreme case of a "just-deposited" material. Conceptually, this would indicate that the soil cannot retain any significant amount of pollutant, the latter being able to move about the grid in a manner reminiscent of hockey pucks on ice. Making the questionable assumption that the weathering half-times described by Langham, Anspaugh and others apply to all erodible material and not just the resuspendible fraction still does not relieve the modeler of the need of postulating mixing depth h, the magnitude of which would be such as to make the results "reasonable," but cannot be known "a priori" in the strictly analytical sense.

The other alternative offered by Travis to account for weathering was to make the thickness h time-dependent, and increasing it as the pollutant penetrates further into the soil, with time. However, as shown previously, this penetration occurs very rapidly during the initial time period immediately after deposition and then remains practically stationary. In order for weathering to proceed in a manner similar to that described by Anspaugh, fr_h would have to include a decreasing exponential term $e^{-\lambda t}$, which, h being the variable of concern, would be roughly equivalent to have h increase exponentially, in a manner dictated by $e^{\lambda t}$. This behavior of h would be extremely unrealistic.

The import of the preceding discussion can best be gaged by examining the vertical flux term, F_v. As expressed in the present model, F_v is a function of F_h and friction velocity u *.

$$F_v = F_h \left(\frac{C_v}{u_t^{*3} C_h}\right) \left[\left(\frac{u^*}{u_t^*}\right)^{p/3} - 1\right] \qquad (190)$$

where

C_v, C_h = constants, defined by Gillette to be 2×10^{-10} and 10^{-6}, respectively.

P = mass percent of suspendible particles

$$= 100 \; \frac{\rho \Delta x \Delta y \; h \; f \; + m_s}{\rho \Delta x \Delta y \; h + m_s + m_n}$$

where f = mass fraction of suspendible soil particles

The mass fraction of suspendible contaminant particles in the vertical flux can be calculated by the ratio

$$fr_v = \frac{m_s}{\rho \Delta x \Delta y \; h \; f \; + \; m_s} \qquad (191)$$

By following the development of $F_v fr_v$ from equations (188) through (191) and the intervening discussion, it can be seen that the weathering effect on resuspendible particles is meant to be incorporated by means of a similar weathering effect on the total erodible mass and horizontal flux F_h, F_v being proportional to F_h. Since Travis' model does not provide, apparently, any efficacious or realistic way of achieving this time dependence, it would follow that the weathering effect cannot be included, at present, in the model as described (Travis, 1975). This represents a drawback of the present model.

Using the vertical flux F_v as a source term for a "Gaussian Puff" model of an instantaneous point source diffusing in three dimensions, the concentration of pollutant at any position (x,y,z) may be obtained by

$$C(x,y,z,t) = \frac{F_v fr_v \Delta x \Delta y \Delta t \; a_{SP} (2\pi)^{-3/2}}{\sigma_x \sigma_y \sigma_z} \, e^{\left[-\frac{(x-x_c)^2}{2\sigma_x^2} - \frac{(y-y_c)^2}{2\sigma_y^2} - \frac{(z-z_c)^2}{2\sigma_z^2} \right]} \tag{192}$$

where

$C(x,y,z,t)$ = concentration of pollutant, μci/cm^3
$\sigma_x, \sigma_y, \sigma_z$ = dispersion coefficient, cm
x_c, y_c, z_c = position of puff center, as cloud moves downwind

Multiplying this equation by $1/fr_v a_{SP}$ will provide the total soil concentration in air at the corresponding locations. Travis estimated that ground level concentrations from this Gaussian Puff model should be correct within a factor of 3.

As the cloud drifts downwind, material is lost from it at a rate determined by the deposition velocity V_d. At present, the model assumes this deposition velocity to be equal to the terminal velocity, V_g.

$$V_d \simeq V_g = \frac{D^2 g}{18\mu} (\rho_p - \rho_a) \tag{193}$$

where V_g = gravitational settling velocity, cm/sec
D = particle diameter, cm
g = gravitational constant,
= 980.665 cm/sec^2 at sea level.
ρ_p = density of particle, gms/cm^3
= 11.4 gm/cm^3 for PuO_2
ρ_a = density of air, gms/cm
= 0.0012 gms/cm at 20°C, 1 ATM
μ = viscosity of air, gm/cm-sec
= 0.000181 gms/cm-sec at 20°C, 1 ATM

As Travis pointed out, this equation is in error when small particles are considered, due to the influence of surface and wind characteristics on the deposition process. Consequently, the model may miscalculate air concentrations of pollutant, particularly when particles of respirable size (<3 μm) are considered.

It should be recalled that Travis's model prime intent was the description of contaminant redistribution, not that of resuspension. Consequently, and in view of the lack of proper weathering terms and deposition velocities applicable to small particles, the conclusion must be made that the model in present form can best be used for short times after deposition, provided that the mass fractions of suspendible and non-suspendible pollutant can be accurately identified, and for particles above a certain, as yet unidentified, minimum size.

CONCLUSIONS

Having reviewed the salient characteristic of each model, commented on possible drawbacks, and added interpretations that may be, hopefully, of some assistance to the modeler, several generalizing statements appear to be in order.

In viewing model disadvantages, it becomes apparent that all the models discussed have a common characteristic - that of failing to identify, at a realistic (finite) source, the progressively diminishing portion of the contaminated soil that may be resuspended.

In gaging models advantages, the possible specific applications of each model must be considered. Suggesting the use of one model to the exclusion of all others is an arduous task, inasmuch as each model has features to recommend it, originating from the conditions by which the model was developed and the purpose of the model development. The simplest models should have greater applicability, whereas more complex models should gain in accuracy. In describing the relevant features of each model and conditions for the model use, the author has opted for a tabular presentation, suggesting the conditions for "best" applicability of each, with the qualification that these conditions are not restrictive (Table 15).

The intervening Table 14 separates the various models into groups, on a somewhat arbitrary basis, depending on what derived parameters, principles, or influencing conditions were applied of greatest significance in the model, in the author's interpretation.

TABLE 14. BASIC GROUPING OF RESUSPENSION MODELS, ACCORDING TO MAIN CONCEPTS EMPLOYED OR DERIVED FROM THEIR USE

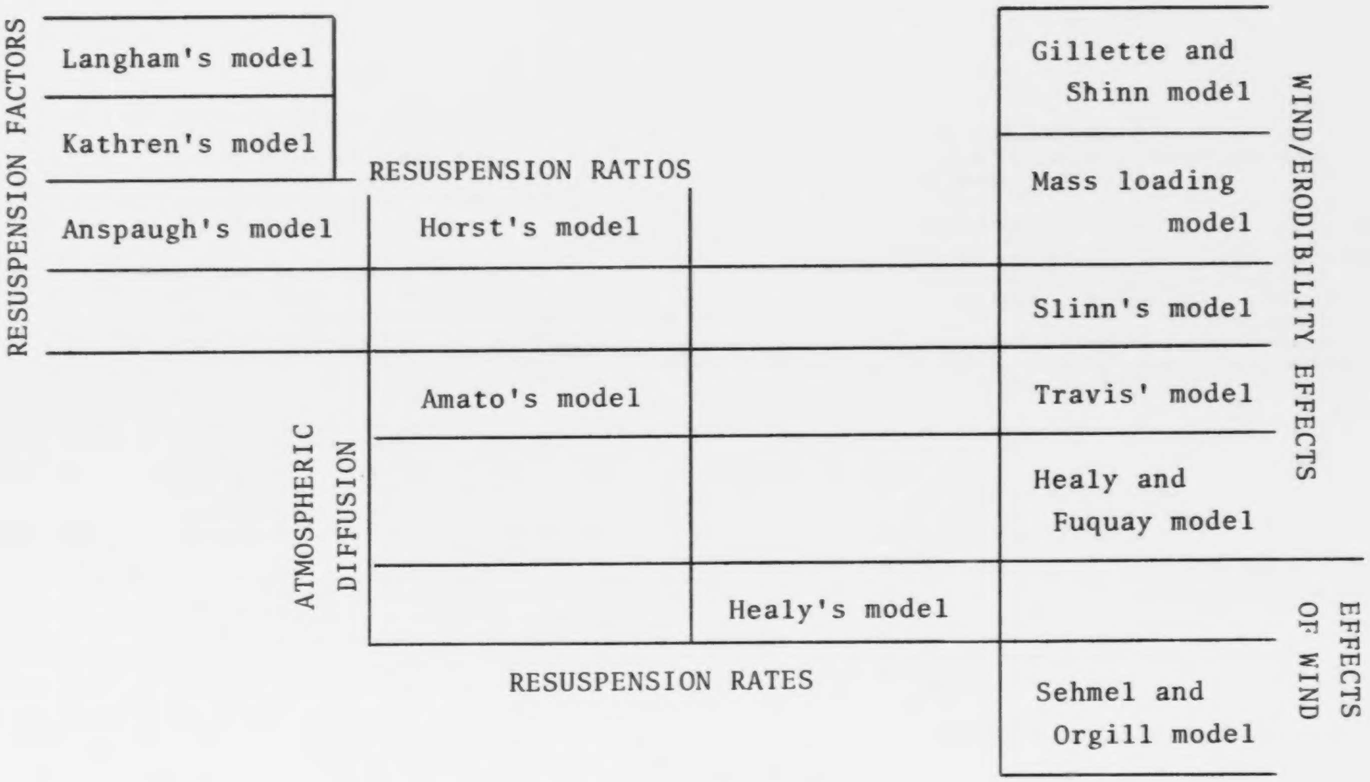


TABLE 15. MAIN FEATURES OF RESUSPENSION MODELS AND CONDITIONS TO WHICH THEY ARE BEST APPLICABLE

Model	Main Features of Model	Conditions to which Model is best applicable
Langham's Model	The airborne concentration of pollutant C(t) (time dependent) is calculated through use of a "resuspension factor" R_F (constant) and the "surface contamination available for resuspension" Sa(t) (time dependent), expressed as an exponential decay function, with an initial value $S_a(0)$, the rate of decay being set by an "exponential coefficient," product of time t and an "attenuation coefficient" λ_A (constant)	1. Uniform distribution of pollutant 2. over large (ideally "infinite") areas 3. for relatively fresh contamination 4. for short time spans For use with $R_F = 10^{-6}m^{-1}$, 5. under conditions of moderate activity (Langham's suggested value 6. in lightly vegetated areas
Kathren's Model	The airborne concentration of pollutant C(t) (time dependent) declines exponentially at a rate set by a coefficient which is product of time t and an "air concentration attenuation factor" λ_A (constant) The initial value of airborne concentration C(0) is related to the surface concentration S_a (constant) through a "resuspension factor" R_F (constant)	Same conditions as above 1 through 4 For use with $R_F = 10^{-5}m^{-1}$(value used by Kathren), add 5 under conditions of extreme activity.
Anspaugh's Model	The airborne concentration of pollutant C(t) (time dependent) is related to the "total concentration per unit area" [regardless of depth] S_a (constant) through a "resuspension factor" $R_f(t)$ (time dependent) expressed as an exponentially declining function, with an initial value $R_f(0)$ (constant) and a rate of decline given by exp $[-0.15/\sqrt{day} \times \sqrt{t}]$ until a final value is reached of $R_f(\infty)$ (constant)	Conditions 1, 2, and 6 above, plus 3. Following an "instantaneous" release or release of short duration 4. for times $t \geq t_r$ - 4 days (100 hours) where t = "model" time t_r = time since release 5. for short time spans following deposition, i.e., for fresh contamination 6. for aged contamination (where $R_f(t)$ $R_f(\infty)$ $10^{-9}m^{-1}$) (little documentation exists for $R_f(t)$ behavior during "intermediate" periods)
Sinn's Model	The resuspension factor R_f, calculated on the basis of several measureable parameters, expressed as soil erodibility E, fraction of horizontal flux at samples height, r mean wind velocity $\bar{u}$, depth to which contaminant has penetrated into the soil, s	1. Uniform distribution of pollutant 2. for aged sources 3. for contaminated areas of finite size, A_S. 4. at distances downwind from the contaminated area such that $\sigma_y \sigma_z \cong A_S$, contaminated area.

TABLE 15. MAIN FEATURES OF RESUSPENSION MODELS AND CONDITIONS TO WHICH THEY ARE BEST APPLICABLE (Continued)

Model	Main Features of Model	Conditions to which Model is best applicable
Mass Loading Model	The airborne concentration of contaminant C, product of "average mass loading" AML, times the specific ground activity A	1. For aged contamination 2. for specific land uses 3. over large areas
Sehmel and Orgill Model	The airborne concentration of contaminant C expressed as function of wind velocity $\bar{u}$ elevated to the 2.1 power	Strictly applicable to conditions and locality where model was obtained 1. At Rocky Flats, Station S-8 2. for West and Southwest winds
Gillete and Shinn Model	The "dust flux" F and concentration C, at 1 meter height, expressed as functions of friction velocity u_* elevated to a power γ (empirically determined), and a "reference dust flux" F_o (at $u_* = 1$ m/sec) Both F_o and γ are represent graphically, as functions of soil erodibility index I (see, however, present author's discussion, eqn (75) and following)	In a strict sense, the model predicts dust fluxes and concentrations, applicable to contaminants if a proportionality between dust and contaminant concentrations were established, in which case the model would best be applicable under the following conditions: 1. over large areas 2. for aged contamination 3. for soils of low erodibility index I
Healy and Fuquay Model*	The airborne concentration of resuspended contaminant C(x,y,0) (location dependent) is calculated through application of diffusion, depletion and source expressions, described below: A source rate, Q function of mean wind velocity u, related to: a surface concentration S_a (constant), of a given contaminant of density ρ (constant), present as particles of a physical diameter d (constant), through a "pickup coefficient" K (constant) Source depletion rates, Q(x)/Q, function of wind velocity u, distance from source x, Sutton diffusion coefficient C (constant), atmospheric stability n (constant) and a "coefficient of deposition" Λ (constant) Atmospheric diffusion equations, C(x,y,0) (location dependent), function of x, c, n, u (already mentioned above), the "crosswind distance" y, and a "reflection factor," R (constant)	1. For contamination sources of specific shapes, as developed by Healy and Fuquay: a. point sources b. line sources of great (ideally "infinite") length c. areal sources of "infinite" crosswind extent but finite along wind direction d. large (infinite") areal sources, 2. with surface contamination distributed uniformly, 3. with contaminant particles of a given diameter, 4. Sutton coefficients such that $C_z = C_y$, 5. at short distances from the ground. 6. for relatively small wind velocities, 7. for slight soil movement.

TABLE 15. MAIN FEATURES OF RESUSPENSION MODELS AND CONDITIONS TO WHICH THEY ARE BEST APPLICABLE (Continued)

Model	Main Features of Model	Conditions to which Model is best applicable
Healy's Model	The airborne concentration of resuspended contaminant $C(x,y,z)$ (location dependent) is calculated through application of atmospheric diffusion, depletion and source expressions as described below: Source rates Q related to surface contamination S_a (constant) through a "resuspension rate" R_R (constant) Source depletion rates, $Q(x)/Q$ function of the distance from source x the Sutton diffusion coefficient C_z (constant) the average wind velocity u the atmospheric stability n (constant) Atmospheric diffusion equations, function of the variables x, C_z, n, u (mentioned above) in addition to: the "crosswind distance" y the elevation z the Sutton diffusion coefficient C_y (constant) the "reflection factor" R (constant)	1. For contamination sources of specific shapes, as developed by Healy: a. point sources b. line sources of great (ideally "infinite") length c. areal sources of "infinite" crosswind extent but finite along wind direction d. large ("infinite") areal sources e. line sources with a Gaussian distribution of contaminant 2. with surface contamination distributed uniformly (excepting 1e above) 3. at "ground level" for cases 1c, 1d, and 1e 4. directly downwind from peak ground concentration in case 1e 5. for particles not much greater than 1.5 micrometers (physical diameter)
Horst's Model	The airborne concentration of resuspended contaminant $C_r(t)$ (time dependent) is calculated by applying a resuspension factor R_F(constant) to the total contamination per unit area $S_a(t)$ (time dependent), obtained through solution of two differential equations describing the rates of change of the two quantified indicated below: A surface contamination $S_a(t)$ (time dependent) affected by three terms, as follows: 1. a "rate of resuspended contaminant deposition" related to resuspended contaminant concentration $C_r(t)$ (time dependent) [itself a function of the surface contamination $S_a(t)$ and a "resuspension factor" R_F (constant)], through a "deposition velocity" V_d (constant) 2. a "rate of surface contaminant soil fixation," related to the surface contamination $S_a(t)$ through a "fixation rate" λ (constant) 3. a "rate of surface contaminant resuspension," related to the surface contamination $S_a(t)$ through a "resuspension rate" R_R (constant) A total contamination per unit area $S_A(t)$, (time dependent) affected by two terms, as follows: 1. a "rate of resuspended contaminant deposition" (with the same relationships mentioned in 1) above) 2. a "rate of surface contamination resuspension" (with the same relationships mentioned in 3) above) Solving these two equations results in a now explicit time dependence, t	1. Freshly deposited contamination 2. uniformly distributed 3. over large areas, 4. for low wind velocities

TABLE 15. MAIN FEATURES OF RESUSPENSION MODELS AND CONDITIONS TO WHICH THEY ARE BEST APPLICABLE (Continued)

Model	Main Features of Model	Conditions to which Model is best applicable
Amato's Model	A computerized model, calculating the "resuspension ratio" R_{r/d_i} as the ratio of airborne concentrations in an interval of finite width and "infinite" length i due to "resuspension" of material deposited in preceding intervals, C_{r_i} and to resuspension from the originally contaminated ground source interval, C_{d_i} The "resuspended" airborne contamination in interval i, C_{r_i} is obtained by summation of contributions from preceding intervals j, each providing at interval i an additional concentration $C_{s_i}^{j} S_j / \rho h$ i.e., the airborne soil contamination at i from interval j, $C_{s_i}^{j}$ weighed by interval j's soil contamination S_j, the depth of penetration of contaminant into the soil h, and the density of the soil ρ, The airborne soil concentration at interval i suspended from interval j, $C_{s_i}^{j}$ is obtained by means of a diffusion equation, including factors such as resuspension flow rate q^r distance between intervals j and i, x Sutton coefficient C_y Sutton coefficient C_z stability coefficient n deposition velocity V_d wind velocity u elevation z The soil contamination in interval j, S_j is obtained as summation from preceding intervals k, of the soil contaminations in k from original source contamination S_1, weighed by a resuspension flow rate q^r (mentioned previously), a "fixation rate" λ, and constants obtained from the particular solutions A_j^{j-k}, to differential equations expressing rates of change of S_j, including h, ρ, q^r, λ (mentioned previously) in addition to a deposition rate coefficient D_j^k and a "time of flight" from k to j T_j^k The "deposition rate coefficient "between k and j, D_j^k is obtained as function of q^r, h, ρ, V_d, x, C_z, C_y, n, u and, in addition, a "reflection factor" R The "time of flight" between j and k, T_j^k is a function of the length of intervals L_n the number of intervals between k and j, n and the wind velocity u The resuspended airborne contamination at i, direct from source "1", C_{d_i} with $C_{s_i}^{j}$ in the diffusion model being replaced by $C_{s_i}^{1}$ with S_j being replaced with S_1	1. For aged contamination (implying intimate association with soil) 2. uniformly distributed with depth, 3. the depth not exceeding top centimeter of topsoil, 4. the topsoil consisting of material able to move in suspension, but not in saltation or creep, i.e., extremely fine material, 5. uniformly distributed over the soil surface, 6. initially, over an area finite along wind direction and of infinite extent in the "crosswind" direction.

TABLE 15. MAIN FEATURES OF RESUSPENSION MODELS AND CONDITIONS TO WHICH THEY ARE BEST APPLICABLE (Concluded)

Model	Main Features of Model	Conditions to which Model is best applicable
Travis' Model	A computerized model, calculating airborne concentration C(x,y,z,t) (space, time dependent) over any one grid cell of area x y of a two-dimensional grid system, as function of the spatial coordinates x y z the time t the dimensions of the grid cell Δx Δy the specific activity of the soil a_{S_p} of the mass fraction of suspendible contaminant in the vertical flux fr_v [itself a function of the mass of suspendible contaminant M_s, the depth to which the contaminant has penetrated into the soil h, the mass fraction of suspendible contaminant in the soil f, the density of the native soil ρ , and the area of the grid cell, $\Delta x \Delta y$], and "the vertical mass flux" F_v [the latter a function of the friction velocity u^*, the threshold surface friction velocity u_t^*, the correlation constants C_v C_h the mass percent of suspendible particles, P and the "horizontal mass flux" F_h] The "horizontal mass flux" F_h, is a function of the effective surface friction velocity u_e^* [function of observed surface shear τ, surface soil moisture content α, density of surface air ρ_a] a reference surface friction velocity u_r^* [function of a reference surface shear τ_r, density of surface air ρ_a (mentioned above)] the already mentioned threshold surface velocity u_t^* the soil erodibility fraction x [function of soil erodibility index I, amount of regetative residue R, surface roughness K, and two constants a b The mass percent of suspendible particles P depends on parameters ρ, Δx, Δy, h, f (already mentioned above) and the mass of non-suspendible contaminant M_{NS} Redistribution of contaminant along the ground is introduced through the "mass fraction of contaminant in the horizontal flux" fr_h, dependent on the same parameters that affect fr_v, plus the "mass of non-suspendible contaminant" M_{NS} Material loss from a cloud moving downwind is related to V_g, gravitational settling velocity equated to "velocity of deposition" V_d, function of particle diameter D, gravitational constant g, density of particles ρ_p, density of air ρ_a, viscosity of air	1. For aged contamination, 2. with resuspendible fraction consisting of particles not over 20 μm in diameter, 3. but sufficiently large to preclude turbulent transfer effects over-shadowing terminal settling velocities, 4. not necessarily uniformly distributed, 5. over a large readily partitionable into planar cells, 6. with a recognizably uniform contamination in each cell An alternative to condition 1) is in applying model to 1a) fresh sources 1b) for short time periods

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PARAMETERS FOR ESTIMATING THE UPTAKE OF TRANSURANIC ELEMENTS BY TERRESTRIAL PLANTS

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PREFACE

The Office of Radiation Programs of the U.S. Environmental Protection Agency carries out a national program designed to evaluate population exposure to ionizing and non-ionizing radiation, and to promote development of controls necessary to protect the public health and safety. This literature survey was undertaken to assess the available information of parameters for estimating the uptake of transuranic elements by terrestrial plants. Readers of this report are encouraged to inform the Office of Radiation Programs of any omissions or errors. Comments or requests for further information are also invited.

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INTRODUCTION

This report summarizes information from the literature concerning parameters which can be used to estimate the transport of transuranic elements through plants to man. The scope of the report is limited to parameters for estimating the concentrations of transuranics in terrestrial plants based on activity concentrations in soil and air.

There is only a limited amount of information specifically concerning plant uptake of transuranics. In many instances it has been necessary to use information based on other elements, which interjects additional uncertainties due to the variance in physical and chemical characteristics of these elements versus the transuranics. Furthermore, most of the transuranic data relates to plutonium; thus, this report focuses on plutonium.

Brown (1975) presents a bibliography of information concerning plant uptake of americium. Americium has only received cursory coverage in this review, although consideration has been given to the differences between americium and plutonium in plant uptake. Differences in the mobility and uptake of plutonium-238 and plutonium-239 are discussed. Papers concerning deposition and retention of plutonium on reindeer lichens have been excluded.

Plant uptake results from root uptake and deposition of contamination on above ground surface areas of the plant. Although deposition or fallout on the plant may not actually be taken into the plant tissue structure, Romney et al. (1975) and Hanson (1975) note that it may be tightly bonded to the plant microstructure and become essentially indistinguishable from material in the plant tissue. Deposition on plant surfaces occurs from both the initial contamination cloud and resuspension of contaminated soil.

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SUMMARY AND CONCLUSIONS

The limited amount of available information is not adequate to select precise parameters for estimating concentrations of plutonium in vegetation due to root uptake and foliar deposition. The existing data base is essentially non-existant for some parameters, and shows significant variations for other parameters. Furthermore, much of the existing laboratory-generated data is not directly applicable to the normal geographical and climatological field conditions.

There is a significant variance in the estimates of the particle size distribution for airborne plutonium. Estimates of aerodynamic mean diameters appear to vary from sub-micrometer to about 10 μm diameter particles. For given particle size distributions, there are uncertainties in the deposition velocity, vegetation interception factors, and retention parameters. Furthermore, much of the experimental data appears to be for particle-size distributions significantly larger than those expected from normal nuclear reactor fuel cycle plant releases, worldwide fallout, or resuspension. There is the additional unknown feature in that specific plant deposition parameter work has not been done with plutonium.

The plant deposition parameter information is summarized in Table 1. These data imply a deposition interceptor factor (F, pCi on vegetation per unit area subtended by the vegetation, per pCi per unit area of ground) of about 0.2. Although the measurement of this parameter is time dependent, the time after deposition is generally not indicated in these studies. It appears that the weathering half-life is short (hours to a day) during the initial deposition period.

There is a significant range in the initial retention estimates associated with the type of vegetation, and more importantly, a variance associated with the time after deposition when the measurement is made. The intervening wind and precipitation conditions are also of prime importance.

It is suggested that the initial retention and weather half-life data should be treated as sets for each individual study. That is, the individual parameters should not be averaged between studies without a detailed evaluation as to common situations (e.g., time of measurements and climatology). Prudence appears to indicate choice of a weathering half-life of about 30 days for time periods of about a week after deposition. For small plants, not prone to trapping particles, a retention factor of about 20 to 30 percent appears reasonable, but there is considerable uncertainty in the data.

Plant uptake parameters are summarized in Table 2. Hanson (1975) estimates the plutonium uptake by plants to be about 10^{-4}. Others generally categorize it as about $10^{-3}$to 10^{-6} which includes most of the data in Table 2. Romney et al. (1970) and Neubold (1963) present data that show a definite increase in plant uptake with successive crops. Romney's et al. (1970) data indicate about an order of magnitude increase, from 1.9×10^{-5} to 14×10^{-5} over a 5-year period. This increase is generally related to microorganism activity in the soil (Au (1974), and Au and Beckert (1975)) and chelation by organics in the soil (Romney et al. (1970)).

There is some indication that plutonium-238 is more mobile than plutonium-239, but this has received only limited verification. Cline (1967) reported that barley took up 50 times as much americium-241 as plutonium-239. Romney et al. (1974 and 1975) have also reported that americium-241 appeared to be more mobile than plutonium.

Essentially all of the plutonium uptake studies are based on laboratory experiments containing plutonium uniformly mixed throughout the soil volume thereby increasing root contact. Results therefore appear to be unrealistic for natural vegetation where the deposited plutonium is largely limited to the upper 2 to 5 cm of soil, above the natural root mat. Therefore, the laboratory results should be conservative for most natural plant species growing in undisturbed or unplowed land; but, the uniform distribution of the plutonium in the laboratory soil should be representative of farm crops grown on plowed land.

Bloom et al, (1974) and Martin et al. (1974) report data from Romney et al. (1974) indicating plant uptake from Nevada Test Site field studies. A total plant uptake of about 0.3 is inferred. They also estimate a total long-term uptake (20 years) of about 0.3 by exponentially extrapolating Romney's et al. (1970) 5-year study to 20 years. Romney et al. (1974 and 1975) qualify the 0.3 uptake as being from both root uptake and deposition and they emphasize that deposition is the primary contributor, probably by several orders of magnitude.

There appears to be general consensus that deposited plutonium is not taken up (by foliar absorption) into the plant; rather, it is generally immobile. This hypothesis is based on limited information.

DEPOSITION ON PLANT SURFACES

The following parameters are used in the deposition pathway:

- Deposition velocity: This may be given with respect to the ground surface or for vegetation. If the deposition velocity for the ground is used, the plant intercept area or factor must be used.

- Plant interception factor (F): Witherspoon and Taylor (1969) defined $F = WC^\circ/m$,

  where:

  W is the biomass of foliage in grams (dry weight) per square foot of soil surface area (g/ft^2).

  C° is the quantity of radionuclide initially intercepted per gram dry weight of foliage ($\mu Ci/g$).

  m is the quantity of radionuclide deposited per ft^2 of soil surface area.

  Thus, F is the ratio of radioactivity deposited on the foliage to the radioactivity deposited on the ground area inhabited by the foliage. The ratio has no units. The product of the deposition velocity for the ground surface and F is the initial effective deposition velocity for vegetation.

- The initial retention factor, (f), is the fraction of radioactivity originally deposited that remains at some time after deposition. This is usually given for one to two weeks after deposition.

- The weathering or retention half-life ($T\omega$) represents the exponential decrease in the retention of deposited activity after the initial one to two week period.

- The plant biomass of foliage (W) is given as grams dry weight per square meter.

- The fraction of deposited activity that is actually taken into the plant is denoted as (d).

CONSIDERATION OF PARTICLE SIZE

The deposition velocity, plant interception factor, and plant retention of deposited material are all a function of the particle size distribution. The size distribution of airborne particulates is related to the source of release. Particle size distributions vary from the micrometer and sub-micrometer diameter for nuclear fuel cycle plants and worldwide fallout to tens of micrometers for near-in fallout and aged fallout in the soil.

High efficiency particulate air filters (HEPA) are used to minimize releases from nuclear fuel cycle plants. HEPA filters have removal efficiencies of 99.97 percent for 0.3 μm diameter dioctyl phthalate smoke particles; thus, releases from most nuclear installations are assumed to be in the sub-micrometer size range (Burchstad (1967)). Moss et al. (1961) also report mass median diameters of less than 1 μm for airborne plutonium in working areas of a plutonium fabrication plant.

Klement (1965) indicates that particulates from nuclear explosions are generally in the sub-micrometer range; but, they may become attached to other material forming conglomerates of 10 μm or more (Gudiksen and Lynch (1975), and Nevissi and Schell (1975)). Generally, worldwide fallout is classed in the micrometer to sub-micrometer diameter size.

The size distribution of resuspended material is dependent on both its original size and composition, and on the material to which it becomes conglomerated within the soil. Bretthauer et al. (1974) analyzed particles from air samples at the Nevada Test Site (NTS) and observed plutonium-bearing particles from less than 0.5 to 17 μm in diameter. The composition of the particles ranged from plutonium, uranium, and oxygen (several micrometer) to silicate and organic particles (about 10 μm). The geometric mean particle diameter was about 1.5 μm.

Volchok et al. (1972) report data from two studies of airborne particulates around the Rocky Flats Plant in Colorado. The initial study, based on particles on particulate filters, indicated a mean diameter around 10 μm. This study was potentially biased by the lack of analysis sensitivity for particles below 0.5 μm. Results from six cyclone and elutricator samples (run time of about 50 hours) indicated median diameters of about 5 μm.

Tamura (1974 and 1975) reports on the particle size distribution of plutonium in NTS soils. One to ten percent of the activity was found in the 0 to 5 μm diameter soil fraction; whereas, about 60 percent (up to 90 percent for several samples) of the activity was found in the less than 53 μm size. Romney et al. (1975) indicate that the micro-structure of many species of vegetation is adept at capturing particles of these size ranges. The bond between the vegetation and these particles is apparently so tenacious as to make it almost impossible to distinguish the material from that actually taken up into the plant.

Bloom et al. (1974) assume a particle size of 10 μm for plutonium in air, which leads to a value of about 5 cm/sec for the deposition velocity. These values are used in their environmental plutonium model.

Bagnold (1945) and Chepil (1945a,b,c,d) indicate that over 90 percent of the wind movement of soil is by surface creep and saltation. These phenomena occur at heights below 1 meter above the ground surface and are not observed on standard air samples. Surface creep and saltation are connected with movement of soil particles of tens to hundreds of micrometers in diameter. Thus, they include movement of the soil size fraction that contains the majority of plutonium (Tamura (1974 and 1975)). Furthermore, vegetation can retain particles of this size class, (Romney et al. (1975)).

In summary, various investigators recommend a range of particle sizes for airborne plutonium. Particle material from original source terms is generally in the micrometer to submicrometer class. Plutonium in soil (limited data, mostly from NTS) appears to be predominately associated with particles between 20-50 μm in diameter (Tamura (1974 and 1975)). The limited information from Bretthauer et al. (1974) and Volchok et al. (1972) indicate that the mean diameter of resuspended plutonium particles is less than 10 μm, probably around 5 μm. Much of the activity deposition on plants with foliage near the ground, would appear to result from surface creep and saltation associated with the larger diameter particles (10 to 100 μm) versus resuspended material.

DEPOSITION PARAMETERS

Witherspoon and Taylor (1969) present data for simulated fallout on pine and oak trees, using cesium-134 as a tracer. The initial fraction (F) of the simulant fallout (88 to 175 μm diameter particles) intercepted and retained by foliage was higher in the oak tree (0.35) than in the pine tree (0.24). After 1 hour, the broad-leaved oak lost about 90.5 percent of the initial deposition, while the pine loss was only about 10 percent, corresponding to initial retention factors of 0.095 and 0.90, respectively. Weathering half-lives (T_ω) due to wind, rain, and all other environmental factors were determined to be 25 and 21 days for the oak and pine trees, respectively, for the period from 7 to 33 days after initial fallout deposition.

Witherspoon and Taylor (1970) present data for five crops using simulated fallout with rubidium-86 as a tracer. Two sizes of quartz particles (44 to 88 μm and 85 to 175 μm diameter) were used on squash, soybean, sorghum, lespedeza, and peanuts. For the size range 44 to 88 μm, the fraction of fallout initially intercepted (F) ranged from 0.075 for the small-leaved lespedeza to 1.248 for the squash. Interception factors (F) greater than unity were obtained for squash and soybean plants. Such plants, which have bush-like structures, have large exposed surface areas available in many different interception planes. The average fraction intercepted (F) for the smaller diameter particle size range was 0.587, which was about 2.5 times greater than F for the larger particle size range.

Fisher (1966) predicts a theoretical decrease in the deposition velocity on pasture grass with decreasing particle size in the 20 to 0.1 μm range. It would appear that using Witherspoon and Taylor's data for the 44 to 84 μm range would be conservative, but there is limited information on which to base this hypothesis.

Witherspoon and Taylor (1970) also studied particle retention. Losses from plant foliage due to wind removal (during the first 12 hours postdeposition) ranged from 3 to 35 percent (average--21.1 percent) for the 44 to 88 μm particles. During the same period, losses for the larger particle size ranged from 9.5 to 26 percent (average--15.8 percent). For the 12- to 36-hour period postdeposition, losses ranged from 1.2 to 33.5 percent (average--15.4 percent) for the smaller size simulant and ranged from 7.7 to 34 percent (average--21.6 percent) for the larger size simulant. Therefore, during the first 12 hours of postdeposition, when the wind speed averaged 0.5 mph and there was no rainfall, the plants lost an average of 18.5 percent of the initial deposition. This corresponds to an average value of the initial retention factor (f) of 0.815. Losses for the next 24 hours also averaged 18.5 percent. From 1.5 to 7 days postdeposition, the plant retention dropped from 63 percent to about 33 percent. This decrease was largely related to 0.25

inches of rain on the sixth day. Retention dropped to 7.9 percent of initial deposition during the second week, after a heavy rainfall of 1.33 inches. Another intense rainfall caused a sharp decrease in fallout retention to 3.3 percent, which slowly decreased during the remainder of the experiment.

The weathering half-lives ($T\omega$) for the 44-84 μm particles were 26 days for the 14- to 28-day postdeposition period and 84 days for the 28- to 56-day postdeposition period. During the entire 56-day study period, the weather half-lives ($T\omega$) ranged from 2.09 to 272.8 days for the 44 to 88 μm particle size; and 1.33 to 56.5 days for the 88 to 175 μm particle size. This experimental data indicates that large differences in initial interception existed between plant species for different particle size distributions, but that these differences become insignificant after about 1 week of exposure to environmental influences such as wind and rain.

Witherspoon and Taylor (1970) report values for the activity per dry gram of foliage, per activity per unit area of soil for the five crops. These values range from 0.01 to 0.2 ft^2/g (10 to 200 cm^2/g). The biomass values (W), in grams of dry foliage per square meter of soil (dried at 100°C for 24 hours), ranged from 20 for lespedeza to 120 for soybeans. These data are for 6-week-old plants at the time of deposition. The plants were planted the last of May in the Oak Ridge, Tennessee, area. All of the foregoing values are summarized in Table 1.

Concentrations of iodine-131 and strontium-89 on plants contaminated by fallout from Project Sedan at the Nevada Test Site were reported by Martin (1965). Examination of the fallout deposited on foliage indicated that most of the activity was due to particles less than 5 μm diameter, with virtually no retained particles greater than 44 μm diameter. The observed effective half-lives for iodine and strontium on the vegetation corresponded to weathering half-lives ($T\omega$) of 17 and 28 days.

Russell (1965) presents a review of interception and retention of airborne material by vegetation. Based on data from Milborn and Taylor (1965) concerning strontium-89, Russell concludes that on the average nearly one-quarter of the deposited fallout material is initially held on edible leaf tissues. An equal quantity may be associated with the basal tissues. The studies of Milbourn and Taylor also indicate that 50 percent of the radioactivity present on the edible herbage per unit area is usually lost in about 14 days. The fraction of initially deposited fission products lost from cabbage plants in a 28-day period ranged from 0.83 for cesium-137 to 0.90 for ruthenium-106 (Middleton and Squire (1961)). Data were also presented which indicated that washing cabbage leaves in water could remove from 10 to 36 percent of the deposited contamination (average of 24 percent). Middleton and Squire also concluded that the extent of radionuclide absorbtion into leaves was of little importance

TABLE 1. SUMMARY OF PLANT DEPOSITION AND RETENTION PARAMETERS

Reference	Plant Interception Factor F	Foliage Biomass (g,dry/m^2)	Initial Retention Factor	Weathering Half-Life (Days)	Half-Life Pertinent Period(Days)	Comments
Witherspoon & Taylor (1969)	0.35		0.095	25	7 to 33	88 to 175 μm particles, oak trees
Witherspoon & Taylor (1969)	0.24		0.90	21	7 to 33	88 to 175 μm particles, pine trees
Witherspoon & Taylor (1970)	0.075 to 1.2	20-120	<0.1@ 2 wk	26	14 to 28	44 to 88 μm particles, 6-wk old plants
Witherspoon & Taylor (1970)		20 lespedeza	∿0.8@ 12 hr			
Witherspoon & Taylor (1970)		120 soybeans	∿0.3@ 1 wk			
Levin et al. (1970)	0.25	540 fruits		14		
Levin et al. (1970)		3500 leaves				
Martin (1965)				17		Fallout, I-131
Martin (1965)				28		Fallout, Sr-89
Russell (1965)			∿0.5			
Russell (1965)			∿0.17			Cabbage, Cs-137
Russell (1965)			∿0.10			Cabbage, Ru-106
Bloom et al. (1974)	5 cm^2/g			30		F factor divided by biomass.
Bloom et al (1974)	∿ 0.14					Assume biomass of 289g/m^2
Martin et al. (1974)	5 cm^2/g			30		
	∿ 0.14					
Milbourn & Taylor (1965)				14		Project Sedan Sr-89

compared to the retention of activity on the leaves.

Levin et al. (1970) discuss the choice of parameters for dose model calculations for the proposed nuclear applications in the construction of an interoceanic canal. The information they present is oriented to South America and general fallout of mixed fission products. Thus, the information has limited applicability to conditions in the United States. A summary of their parameters is given in Appendix A. Several of the more pertinent values are:

--Fraction of the element in plant edibles which comes from leaves (due to foliar deposition) of 0.05. This value apparently relates to fruit type plants.
--W, biomass of plant edibles of 540 g/m^2 (dry weight). Biomass of plant leaves 3500 g/m^2.
--Growth-rate coefficient for plant edibles of 0.05 day^{-1} or half-life of 14 days.
--Weathering elimination rate for plant leaves of 0.05 day^{-1} or 14-day half-life.
--F, Fraction of fallout intercepted by plant leaves of 0.25.

The weathering elimination rate (λ_ω) was estimated to be 0.05 day^{-1} based on a T_ω of 14 days. The fraction of fallout intercepted by plant leaves was 0.25. For most fission products the fraction in the plant edibles which comes from leaf contamination was estimated to be 0.001. The fraction in plant edibles which comes from the root uptake of contaminants in soil was 1.0. These values are estimates influenced by natural weathering conditions and decontamination due to washing and food preparation.

Bloom et al. (1974) review the literature values to obtain parameters for use in their transport model. Based on fission product fallout data, they postulate the following half-lives:

Half-Life (Days)	Time Increment (Days)
1.4	0 - 5
20	5 -15
30	15 -30
130	30 -60

Bloom et al. (1974) note that such parameters as interception, retention, and retention half-life are dependent on the time after deposition when these factors are measured. Given this, they select an interception factor (units of cm^2/g dry weight) of 5 and a weathering half-life of 30 days. This interception factor (in units of cm^2/g) is equivalent to the unit-less interception factor (F) divided by the plant biomass

(W) in g/cm^2. Bloom et al. (1974) refer to the studies by Miller and Lee (1966) of interception factors (cm^2/g) for fallout from a volcano eruption. Values of the interception factor vary by a factor of two (from 47 to 96 cm^2/g) between low and high (greater than 90 percent) relative humidity. Miller and Lee (1966) note that these values were based on samples collected immediately after deposition. Notation of time after deposition may explain the discrepencies with nuclear fallout data reported by Martin (1965). Martin (1965) reported values from 1.9 to 11.1, with an average of 3.7 cm^2/g. For relative comparison purposes, Bloom's et al. (1974) factor of 5 cm^2/g can be converted to the unit-less factor by assuming a biomass of 289 g/m^2 (Martin et al. (1974)), resulting in an F value of 0.14.

Martin et al. (1974) uses parameters similar to those of Bloom et al. (1974). Several of the recommended parameters are: interception factor of 5 cm^2/g; weathering half-life of 30 days (based on nonvolatile particulates on shrubs); and a desert plant dilution growth rate of 36 g(dry)/m^2-year. A biomass value of 289 g/m^2 is referenced (Bamberg (1973)).

RADIONUCLIDE UPTAKE FROM SOIL BY PLANTS

The uptake of radioactive material from soil by plants is generally expressed as a discrimination factor pCi/g (dry weight) of plant per pCi/g (dry weight) of soil. Variations of this parameter include the discrimination factor between soil and the roots or the edible fruits of the plant. The discrimination factor is also given for uptake from hydroponic solutions. Table 2 summarizes the plant uptake of plutonium studies discussed below.

The depth basis of the soil concentration presents an inherent uncertainty in the discrimination factor. The majority of fallout plutonium is normally found in the top two-to-five cm of soil. Thus a 10-cm depth soil sample will only contain essentially one-half the concentration of plutonium as a 5-cm depth sample (i.e., the 10-cm sample is diluted with uncontaminated dirt). It appears that most discrimination factors are based on a 5-cm depth soil sample.

Several investigators have indicated an increase in the discrimination factor with time (Romney et al. (1970); Price (1973); Francis (1973)). The extent of and reasons for this increase are uncertain. It is generally related to either the chronological increase in depth penetration of plutonium in soil and/or the increased availability of plutonium with time.

The increased penetration is related to alternate freezing - thawing, and wetting - drying of the soil; earthworm activity; agriculture practices; possibly changing plutonium solubilization; and physical translocation downward through the soil by the root hair system of plants (Wildung and Garland (1974)). Wildung and Garland (1974) noted that plutonium from surface soil was translocated down to the roots of barley. This may have special health pathway implications for root crops directly consumed by man.

Chronological increases in the bio-availability of plutonium are related to the natural chelation of plutonium by decaying roots (Romney et al. (1970)). Au (1974) and Au and Becker (1975) indicate significant uptake of PuO_2 by soil microorganisms, specifically Aspergillus niger. Their experiments, conducted at several values of soil acidity, indicate that microorganisms may chronologically increase the bio-availability of PuO_2 microspheres.

TABLE 2. SUMMARY OF PLANT UPTAKE OF PLUTONIUM

Reference	Discrimination Factor (pCi/g dry plant: pCi/g dry soil)	Time of Measurement (Yrs)	Type Plant	Comments
Romney et al. (1970)	1.9×10^{-5}	1	Ladino Clover	Pu-239 from NTS soil
"	4.1×10^{-5}	2	" "	" " " "
"	4.4×10^{-5}	3	" "	" " " "
"	7.1×10^{-5}	4	" "	" " " "
"	14×10^{-5}	5	" "	" " " "
Garland et al. (1974)	4.4×10^{-5}	<1	Barley	$Pu(NO_3)_4$ 100 µCi/g in soil, toxic effects
"	15×10^{-5}	<1	Barley	$Pu(NO_3)_4$ 10 µCi/g in soil
"	1×10^{-7}	1/3	Barley	seeds
Johnson et al. (1972)	200 (roots)		Barley	roots, activity may not have been taken up
"	0.8 to 4×10^{-3} Aerial portion: root			Used PuO_2
"	2×10^{-6} to 10^{-3}			$Pu(NO_3)_4$
"	Av 6.4×10^{-4}			
Jacobson & Overstreet (1948)	10^{-4}	1/365	Barley	$PuO_2{}^{2+}$
"	2×10^{-5}	1/365	Barley	Pu^{4+}
"	4.5×10^{-6}	1/365	Barley	Pu^{3+}
"	0.4 (roots)		Barley	$PuO_2{}^{2+}$
"	0.25 (roots)		Barley	Pu^{4+}
"	0.2 (roots)		Barley	$PuO_2{}^{2+}$
Cline (1967)	Am-241 50xPu-239		Barley	
Cline (1967)	0.003 for Am			Alkaline Ephrata fine sandy loam
"	2×10^{-4}			Pu acid soil
Cline (1967)	1×10^{-4}			Pu alkaline soil
Neubold (1963)	Factor 4 increase	2	Ryegrass	Pu in acid soil
Nishita et al. (1965)	10^{-4}		Ladino clover	Pu Fallout
Rediske et al. (1955)	9×10^{-4}		Barley	Pu^{4+}

Barth (1975), Raabe (1973), Hakonson and Johnson (1973), and Patterson et al. (1974) report data inferring that plutonium-238 is more soluble than plutonium-239, and possibly more biologically available. Raabe et al. (1973) report that the dissolution rate for plutonium-238 dioxide monodispersed particles in an *in vitro* laboratory system was nearly two orders of magnitude greater than for plutonium-239. Hakonson and Johnson (1973) report changes in the plutonium-238 to plutonium-239 ratio for the Trinity Site, New Mexico. Twenty-three years after the nuclear detonation, the plutonium isotopic ratios varied from 0.05 for soil, 0.10 for plants, to 1.0 for mammals. Brown and McFarlane (1975) are conducting experiments with several plant species and soils to determine uptakes for plutonium-238 and plutonium-239.

Hanson (1975) notes that the increased availability of plutonium-238 may result from the chelating action resulting from more intimate contact of plant roots with the plutonium particles (plutonium-238 versus plutonium-239); transport of plutonium by individual cells; or a combination of such mechanisms by which plutonium-238 may be absorbed differently than plutonium-239. The higher specific activity of plutonium-238 versus plutonium-239 is also potentially related to possibly different isotopic effects.

Data indicating differences in the transfer or isotopic ratios for plutonium-238 and plutonium-239 should be critically evaluated. Plutonium-236, which is often used as an analytical tracer may contain plutonium-238 as a contaminant. This error can be corrected by analyzing tracer blanks. Furthermore, sources of purer plutonium-236 are now available. An additional problem results from the similarity of the americium-241 and plutonium-238 alpha energies, 5.49 and 5.50 MeV, respectively. Incomplete separation of americium in sample processing or delays in counting after sample processing (i.e., amercium-241 ingrowth from plutonium-241) can result in erroneously high indications of plutonium-238 content. Generally, these pitfalls are not present, but their potential must be recognized.

Plant uptake of plutonium from soil has been reviewed by Bloom et al. (1974), Hakonson (1975), Hanson (1975), Price (1973), and Francis (1973). The general consensus is that short-term uptake is minimal, but that increased chronological uptake due to natural chelation and other mechanisms presents an uncertain picture and some cause for concern.

Romney et al. (1970) studied the transfer of plutonium-239 from soil to plants for ladino clover. These crops were grown under glasshouse conditions on contaminated soil for five years. Total crop yields increased each year. The plant-to-soil discrimination factor for the first year was 1.9×10^{-5} pCi/g of dry plant per pCi/g of dry soil. The factor increased to 14×10^{-5} for the fifth year, for a five year average of 6.3×10^{-5}. The soil was a one-to-one mixture of Yolo soil and soil from Area 11 of the Nevada Test Site contained in 60-liter containers (filled volume 50 liters), with a surface area of 0.12 m^2. The soil was uniformly contaminated with a plutonium-239 concentration of 1.62×10^{-5} dis/min-g. Romney et al. speculated that some of the yearly increase in plutonium uptake was related to increased development in the root system. They maintained the root system provided more intimate contact of the roots with plutonium. Additional studies showed increased plant uptake of plutonium from soils where DTPA chelating agent was added.

Garland et al. (1974) report results for barley and soybean plants grown in soils containing $Pu(NO_3)_4$. The split-root technique was used to study the uptake and distribution of plutonium in the plant tissue. The distribution of plutonium was determined in the tops and roots of soybeans (Glycine max) after 50 days of growth, and barley (Hordeim vulgare) after 27 days of growth. Slight increases in the total plant uptake were related to increasing the volume of soil in the test plots for both above-ground and root tissues of barley. But the height of the soil column appeared to be a more important variable. Since the plutonium was uniformly mixed in the soil column, increased uptake from a taller soil column probably relates to the increased contact between roots and soil.

Garland's et al. (1974) experiments were conducted with concentrations of 10 and 100 μCi/g of plutonium-239. But the elevated concentration of 100 μCi/g did not result in a marked increase in uptake versus the 10 μCi/g soil. The respective plant uptakes (dry weight, 60°C for 24 hours) for barley were 4.4×10^{-5} and 15.5×10^{-5}. The plants in the 100 μCi/g soil showed toxicity symptoms until the root systems were established in the nutrient solution below the soil. Plants grown in the two concentrations were indistinguishable at the time of harvest. If the observed toxicity is concentration dependent, Garland et al. (1974) indicate that the previously reported results of Wildung and Garland (1974), indicating an inverse relationship between uptake and soil concentration, may have been due to toxic effects on the roots.

Garland et al. (1974) reported that the distribution of plutonium activity in the plant roots of barley averaged 17.1 percent and 4.79 percent (percent of total plant activity) for two different plutonium activity soils (0.05 and 10 μg plutonium per μg soil). Therefore, the average root content was 10.95 percent, with the remaining plant activity in the above-ground parts of the plant (outer sheath, leaf blades and new growth). Both the barley and soybean plant studies indicated that plutonium, once in the plant, was rather mobile, with leaf tissue containing 5 to 10 times the plutonium activity of that in the stem tissue. After 100 days of growth, the barley seed had an activity corresponding to a concentration factor of less than 1×10^{-7} of the soil content.

Johnson et al. (1972) reported the results of an experiment to test the active transport of plutonium by plant roots. Barley plants were grown in a nutrient media containing soluble $^{239}Pu(NO_3)_4$. The barley plant roots were removed at various time intervals, washed and analyzed for plutonium-239 content. A tentative concentration factor from solution to roots of approximately 200 was observed. Although the roots were washed after removal from the nutrient media, it is possible that much of the plutonium was only associated with exterior surface contamination of the roots and was not assimilated by the roots.

Johnson et al. (1972) conducted another experiment where barley plants were allowed to mature and the root and aerial portions were removed and separately analyzed. The ratio or fraction of the concentration of plutonium in the aerial portion of the plants, as compared to the root portion of the plants, ranged from 0.8x10^{-3} to 4x10^{-3} (average of 2x10^{-3})for plants grown in PuO_2 solutions. The ratio for plants grown in $Pu(NO_3)_4$ solutions ranged from 2x10^{-6} to 1x10^{-3}, with an average of 6.4x10^{-4}. These ratios are similar to those reported by Romney et al. (1970). This indicates that either the plutonium was not taken up into the roots, or there is a significant discrimination factor preventing the transfer of plutonium from roots to the above-ground parts of the plant.

Menzel (1965) reviewed the literature concerning the soil-plant relationships of radionuclides. This review was limited to experiments where the radioactivity was in a soluble form when added to the soil and where radionuclide concentrations were low enough so that there were no toxic effects. In summary, Menzel classed plutonium in the bottom of the lowest category (that is, less than 0.01 (ratio of dry weights of plant and soil activity)).

Francis (1973) reviewed the mobility of plutonium in soil and its uptake by plants. The following items are based on Francis' review:

1. Jacobson and Overstreet (1948) studied the translocation of plutonium in barley (one of the original plutonium plant uptake studies of barley plants in calcium-bentonite clay suspensions). Over a 24-hour period, the fractional translocations to leaves, were 10^{-4} for PuO_2^{2+}, 2x10^{-5} for Pu^{4+}, and 4.5x10^{-6}for Pu^{4+}. The respective values for the roots were 0.4, 0.25, and 0.2.

   Rediske et al. (1955) noted the discrimination factor (ratio of dry weight of aerial portion of plants to soil) increased from 10^{-4} to 10^{-3}, with pH changes of 7 to 4.

2. Wilson and Cline (1966) studied the uptake of plutonium-239 by barley from three soils. The soils were Ephrata fine sandy loam, a slightly alkaline soil; Milville silt loam, a calcareous soil; and Cinabar silt loam, a moderately acid forest soil. Plutonium uptake from the acid soil was more than three times greater than from the calcareous soil. A 0.1N nitric acid solution removed 0.64 percent of the plutonium, approximately one-thousand times more than that taken up by barley. This shows that common soil extracting methods do not provide a reliable indication of potential plant uptake.

3. Cline (1967) reported that the uptake of americium-241 into foliar portions of barley was fifty times that of plutonium-239. The barley was grown in Hoagland's nutrient solution.

4. Unpublished work of Buckholz et al. does not show a chronological increase in the discrimination factor for alfalfa after four years of growth. The study was conducted in a plutonium contaminated soil associated with the 1966 Palomares Spain accident. This is at variance with the results of Romney et al. (1970).

Price (1973) reviewed several studies concerning plant and animal uptake of plutonium. The following studies were not reported by Francis (1973):

1. Nishita et al. (1965) studied the uptake of fallout plutonium in ladino clover (*Trifolium respens L.*). The discrimination factor was 10^{-4} (μCi/g plant per μCi/g soil, probably dry weights).

2. Rediske et al. (1955) noted that Pu^{4+} becomes associated with root surfaces exposed to culture solutions. The quality of sorption to root surfaces is linear with respect to concentration of the solution, whereas, the leaf concentrations had a curvilinear relationship. The uptake into shoot tissues of tumbleweed from solution cultures was slightly less than for beans, barley, or tomatoes. The discrimination factor for barley was 9×10^{-4}, based on the Neubauer test. This was considered to be an overestimate for what would be expected under field conditions.

3. Cummings and Bankert (1971) used culture pots for plutonium-238 uptake studies for nine soils. The results for plutonium-238 were lower than those for cerium-144 and promethium-147. The fractional uptake (total activity in plants divided by total soil activity) for plutonium varied from 7 to 280×10^{-8}.

4. Cline (1967) reports discrimination factors of 0.003 for americium-241 from alkaline (Ephrata lime sandy loam) and acid (Cinabar silt loam) soils. The plutonium factors were $2x10^{-4}$ (acid soil) and $1x10^{-4}$ (alkaline soil).

5. Neubold (1963) reported that although plutonium uptake by perennial ryegrass (Folium perenne L.) was low, it did increase over a 2-year study span, for several different soils. There was a 4-fold increase for an acid soil.

Price (1973) indicates the following ranking for decreasing uptake by plants from soil: curium, americium, and plutonium. Neptunium uptake probably resembles that of plutonium.

Hakonson (1975) reviewed pathways for plutonium into terrestrial plants and animals. Several investigators have noted higher plutonium concentrations in native grasses than for forbs, shrubs, or trees (e.g., Hakonson and Johnson (1973) and Whicker et al. (1973)). This may be related to the morphological structures of the plants and their ability to intercept and entrap airborne material. Russell (1966) has noted that the heads of grains serve as an excellent trapping device for deposited material. On the other hand, the physical structure of root systems of grasses and their position within the soil/plutonium profile may be favorable for root uptake of plutonium by grasses.

Bloom et al. (1974) present an environmental transport model with associated parameters. A soil to plant discrimination factor of 0.313 (pCi/g of dry vegetation per pCi/g of wet soil) is recommended. This factor is based on the indications of increases of plant uptake with time (e.g., Martin's et al. (1974) estimates from data from Romney et al. (1974)). The Romney et al. (1974) values are based on results from the Nevada Test Site, 20 years after deposition. Martin et al. (1974) further justified the value of 0.313 by estimating the uptake at 20 years from Romney's et al. (1970) data. In essence, Martin et al. (1974) exponentially extrapolated Romney's et al. (1970) 5-year study to 20 years.

Romney et al. (1974 and 1975) emphasize that the high discrimination factors are not solely related to root uptake, rather, they are a result of deposition with limited root uptake. Romney et al. (1975) estimate the root uptake to be 10^{-3} to 10^{-4}. Thus, it appears Bloom's et al. (1974) and Martin's et al. (1974) assumptions are in error.

COMBINATION OF DEPOSITION AND PLANT UPTAKE

Romney et al. (1974 and 1975) reports data on the total plutonium and americium-241 concentrations in vegetation for several areas of the Nevada Test Site. These data are compared on a pCi/g (dry and ashed) basis to soil concentrations. The vegetation results are based on a sample of foliage, and exclude the root mat. Romney et al. (1975) estimated that for Area 13 only 1/1600 of the plutonium-239 inventory was in the vegetation. The ratio of foliage to soil values averaged about 0.08, and most of the values (several hundred) fell within 0.02 to 0.16.

Romney et al. (1974) reported values for the total amount of plutonium-239 on foliage versus the soil concentration. The preliminary results for vegetation were based on the ashed weight. The average values (pCi/g ashed vegetation per pCi/g dry soil) were 1.0, 1.7, and 5.1 for *Atriplex canescens*, *Atriplex confertifolia*, and *Eurotia lanata*, respectively. The average value was 2.24. Romney's plant-to-soil ratios are apparently all based on the plutonium concentration in the top 5 cm of soil.

Colorado State University (1973) reported data on the plutonium-239 inventory for the Rocky Flats, Colorado, area. The data is based on plant distributions from the Pawnee National Grassland. The soil accounted for 99.464 percent of the plutonium-239 in the top 2 cm of soil. Standing vegetation accounted for 0.058 percent, litter for 0.180 percent, and roots (surface to two cm depth) accounted for 0.298 percent of the total activity on the test plot. Considering the litter as part of the standing vegetation, the foliage would then account for 0.238 percent of the total activity, comparable to the root content of 0.298 percent. The total plant content would be 0.536 percent.

Whicker et al. (1973) reported plutonium concentrations for various terrestrial ecosystems in the Rocky Flats environs. In the top 3 cm of soil, fifty-nine percent of the plutonium-239 was found in the soil fraction of less than 0.5 cm in diameter. Additionally, 39 percent of the soil activity was found to be below the 3 cm depth. Two-tenths percent was associated with the surface litter and detritus, 1.3 percent with roots, and 0.06 percent with standing vegetation. Considering the litter as part of the standing vegetation, the foliage would then account for 0.26 percent of the total activity, compared to the root content of 1.3 percent. The total plant content is 1.56 percent, corresponding to a concentration factor of 0.0156, due to both deposition and soil uptake. The root, foliage, and soil data from Whicker (1973) are summarized in Table 3.

TABLE 3. PLUTONIUM IN VEGETATION AND SOIL

(Whicker (1973))

Plant	Biomass Dry Weight (g/m^2)	Average Conc. In Roots (dpm/g)	Average Conc. In Standing Plant (dpm/g)	Standing Veget. Conc: Soil conc.*
Western Wheatgrass	32	247	30	0.00125
Cheatgrass	10	294	112	0.0467
Prickly lettuce	2	157	13	0.00542
Salsify	9	13	13	0.00542
			Average	0.0015±0.02

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* Vegetation concentration in dpm/g divided by average soil concentration of 2397 dpm/g. Based on soil sample of 0 to 3 cm depth and particles less than 5 mm. Dry Weights.

Schultz et al. (1974) report a proposed study of plant uptake of plutonium and americium. The study will include several soils and several chemical forms of the elements. Results have not yet been published.

REDISTRIBUTION OF ACTIVITY WITHIN PLANTS

There appears to be very little absorption and redistribution of deposited plutonium within plants. There is, however, only a limited amount of published information.

Russell (1965) reviewed several studies concerning interception and retention of airborne material. He concluded that the absorption of deposited material was of limited importance compared to the retention of activity on foliar surfaces.

Aarkrog (1975) studied the uptake of deposited fission products on wheat and barley crops. Radionuclides such as strontium-90, ruthenium-103, and cerium-144 were generally immobile. Whereas, zinc-65, iron-55, cesium-137, cobalt-60, and manganese-54 were more readily translocated to the seeds.

Levin et al. (1970) estimate that only 0.1 percent of the radioactivity in plant edibles comes from the leaves (for relatively immobile elements). Essentially all of the activity in the plant edibles is related to root uptake. The parameters listed in Appendix A (Levin et al. (1970)) are for the interoceanic canal project and are assumed to relate to fruits, nuts, etc., -- not to leafy edibles.

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Appendix A

Parameters for Atlantic-Pacific Interoceanic Canal Model (LEVIN et al., 1970)

Parameter	Parameter Definition	Values Used	Remarks	References
F_A	Average fallout concentration on a watershed, µCi/cm^2	-	These values are classified and thus are not available	-
S_{01}	Initial specific activity of the radionuclide in the fallout, µCi/g element	-	These values are classified and thus are not available	-
f_{43}	Fraction of the element in plant edibles which comes from leaves (dimensionless)	0.05 (P&C)$^{(a)}$	For mobile elements H, P, I, and C	Martin (1969)
		0.001 (P&C)	For all other elements than H,P, I, and C	Martin (1969)
f_{42}	Fraction of the element in plant edibles which comes from the soil (dimensionless)	1.0 (P&C)	For all elements except carbon	Kazmaier (1969)
		0.001 (P&C)	For carbon	Kazmaier (1969)
f_r	Ratio of runoff water to total rain water (dimensionless)	0.9 (P&C)	Infiltration is 10 percent of rainfall reaching ground surface	Charnell et al (1969)
F	Ratio of the amount of radionuclide dissolved in surface water to the total amount present on the soil surface (dimensionless)	4.0×10^{-5} (P&C)	For all elements except hydrogen; calculated from reference data$^{(b)}$	Charnell et al (1969)
		0.3 (P&C)	For hydrogen; calculated from reference data$^{(b)}$	Charnell et al (1969)
r	Average rainfall rate, cm/day	0.636 cm/day (P) 0.596 cm/day (C)	Estimated from mass rainfall curves Estimated from rainfall curves	Charnell et al (1969) Charnell et al (1969)
r_u	Unit rain, cm	2.5 cm (P&C)	Defined by reference$^{(c)}$	Charnell et al (1969)
W_4	Dry biomass of plant edibles, g dry weight/cm^2	0.054 g/cm^2 (P&C)	Calculated from reference data$^{(d)}$	Transeau (1926)
X_{W4}	Fraction of water in plant edibles, g water/ g fresh weight	• 0.70 g/g (P&C)	Estimated average water content from reference data	Wu Leung and Flores (1961)

Appendix A (continued)

Parameter	Parameter Definition	Values Used	Remarks	References
ρ_w	Density of water, g/cm^3	1.0 g/cm^3 (P&C)	Listed in reference	Weast and Selvy (1967)
λ_{B6}	Biological elimination rate coefficient of element from freshwater fish, day^{-1}	0.055 day^{-1} (P&C)	Geometric mean calculated from reference data by method outlined by Bloom et al (1970)	Templeton et al (1969) Brungs (1967) Boroughs et al (1956) Polikarnpov (1966 a&b) Kevern (1966) Wiser and Nelson (1964) Friend et al (1965)
λ_{B7}	Biological elimination rate coefficient of element from animals, day^{-1}	0.1 day^{-1} (P&C)	Geometric mean calculated from reference data	Golley et al (1969)
λ_{B9}	Biological elimination rate coefficient of element from marine fish, day^{-1}	0.02 day^{-1} (P&C)	Estimated from turnover rates for anchoveta	Lowman et al (1970)
λ_g	Growth-rate coefficient for plant edibles, day^{-1} $\lambda_g = (\frac{d\,C_4}{dt})/C_4$	0.05 day^{-1} (P&C)	Arithmetic mean calculated from reference data by method outlined by Bloom et al (1970)	Malavolta et al (1962) Transeau (1926) Jacob and von Uexkull (1963)
k_w	Weathering elimination rate coefficient from plant leaves, day^{-1}	0.05 day^{-1} (P&C)	Calculated from reference data$^{(f)}$	Martin (1965)
f_I	Fraction of fallout intercepted by plant leaves (dimensionless)	0.25 (P&C)	Highest value selected from reference data$^{(g)}$	Nishita et al (1965) Middleton (1960)
W_3	Biomass of plant leaves, g dry weight/cm^2	0.35 g/cm^2 (P&C)	Calculated from reference data$^{(h)}$	Transeau (1926)
F_{AM}	Average fallout concentration on the marine fallout area, µCi/cm^2	-	These values are classified and thus are not available	-
Q	Total amount of radionuclide initially present in the canal channel and rubble, µCi	-	These values are classified and thus are not available	-
F_c	Ratio of the amount of radionuclide dissolved in the canal water to the total amount present in the canal channel and rubble (dimensionless)	4.0 x 10^{-3} (P&C)	For all elements except hydrogen$^{(i)}$	Essington (1969)
		0.3 (P&C)	For hydrogen	Essington (1969)

Appendix A (continued)

Parameter	Parameter Definition	Values Used	Remarks	References
B_c	Net flow rate of water through the canal channel, cm^3/day	2.83×10^{14} cm^3/day (P)	Estimated from reference data	Harleman (1967)
		3.62×10^{11} cm^3/day (C)	Estimated from reference data	Harleman (1967)
H	Horizontal extent of the marine fall-out field in the direction perpendicular to the current, cm	1.85×10^7 cm (P-Pacific side)	Estimated from reference data(j)	Ferber (1968)
		1.20×10^7 cm (P-Atlantic side)	Estimated from reference data(j)	Ferber (1968)
		1.11×10^7 cm (C-Pacific side)	Estimated from reference data(j)	Ferber (1968)
Z	Horizontal extent of the marine fall-our field in the direction parallel to the current, cm	1.11×10^7 cm (P-Pacific side)	Estimated from reference data(j)	Ferber (1968)
		7.4×10^6 cm (P-Atlantic side)	Estimated from reference data(j)	Ferber (1968)
		9.26×10^6cm (C-Pacific side)	Estimated from reference data(j)	Ferber (1968)
K	Turbulent diffusivity in the vertical direction, cm^2/day	1.0×10^6 cm^2/day (P&C)	Estimated from reference data(k)	Pritchard et al (1966)
V_c	Volume of water in the canal channel, cm^3	2.52×10^{15} cm^3 (P)	Estimated from reference data	Harleman (1967)
		2.19×10^{15} cm^3 (C)	Estimated from reference data	Harleman (1967)
V_o	Speed of ocean current, cm/day	1.0×10^6 cm/day (P)	Estimated from reference data(k)	Lowman et al (1970)
		3.0×10^6 cm/day (C)	Estimated from reference data(l)	Lowman et al (1970)

(a) P designates value used for Panama (Route 17). C designates value used for Colombia (Route 25).

(b) The quantity F_w was calculated as follows:

$$F_w = \frac{1}{[1 + (\frac{1-\alpha}{\alpha}K_D)]} ,$$

where α is the fractional soil porosity, 0.3

and K_D is the distribution coefficient of the rainwater between the soil surface and surface water, 1 for hydrogen, and 1.0×10^4 for all other elements.

(c) If the interflow layer has a thickness of 7.5 cm (Odum, 1967) and a porosity of 33 percent, then the amount of rain required to saturate this interflow layer is the unit rain (2.5 cm).

Footnotes for Appendix A (continued)

(d) The quantity W_4 was calculated as follows:

$$W_4 = \left[\frac{\text{dry weight plant edible}}{\text{plant}}\right] \times \left[\frac{\text{No. plants}}{\text{cm}^2}\right]$$

$$W_4 = \left[\frac{\text{216g plant edible}}{\text{plant}}\right] \times \left[\frac{1.0 \times 10^4 \text{ plants}}{\text{acre}} \times \frac{2.471 \times 10^{-6} \text{ acres}}{\text{cm}^2}\right]$$

(e) Reference data described the growth of tropical plants. Pineapple, sugar cane, rice, and bananas were some of the foodstuffs for which growth rate data were reported.

(f) The quantity k_w was calculated as follows: $k_w = \frac{0.693}{T_e}$

where T_e is the environmental half-life of a radionuclide on leaves.

T_e was assumed to equal to 14 days for all radionuclides on fallout-contaminated plants in humid regions.

(g) The relative percentage of fallout intercepted by plants in the environs at NTS (Nishita et al, 1965), from 8 percent to 15 percent at the Maralinga Test Site for close-in and far-out fallout, respectively, (Nishita et al 1965), and up to 25 percent retention near NTS (Middleton, 1960).

(h) The quantity W_3 was calculated as follows:

$$W_3 = \left(\frac{\text{dry weight plant leaves}}{\text{plant}}\right) \times \left(\frac{\text{no. plants}}{\text{cm}^2}\right)$$

$$W_3 = \left(\frac{\text{140 g leaves}}{\text{plant}}\right) \times \left(\frac{1.0 \times 10^4 \text{ plants}}{\text{acre}} \times \frac{2.47 \times 10^{-6} \text{ acre}}{\text{cm}^2}\right)$$

(i) The quantity F_c was calculated for all elements except hydrogen as follows: $F_c = 100\ F_w$.

(j) This quantity is dependent upon the orientation of the fallout patterns over the marine fallout area. As no fallout is expected over the Atlantic side of Route 25, no values are listed for the Atlantic side of Route 25.

(k) This quantity is the value used for both the Atlantic and Pacific sides of Route 17.

(l) This quantity is the value used for the Pacific side of Route 17.

A MODEL TO ASSESS POPULATION INHALATION EXPOSURE FROM A TRANSURANIUM ELEMENT CONTAMINATED LAND AREA

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Introduction

Land areas contaminated with radioactive material will cause radiation exposure when the material is resuspended from the ground and dispersed by wind action to populated areas where it is inhaled or ingested. To assess the overall impact of such a contaminated area on the surrounding population, requires a summing of all individual doses over the entire time the material is capable of exposing the population; this is the environmental dose commitment.

A model is proposed that estimates the population inhalation exposure, as a function of time and distance, that results from a single source of radioactive contamination. This permits the estimation of the environment dose commitment from the inhalation pathway. Population inhalation exposure, as used in this report is the integrated air concentration multiplied by the number of persons exposed to this concentration, and has units of person Ciyr/m^3. A method is described to convert population inhalation exposure to an environment dose commitment, which then may be used to estimate the number of potential health effects.

The model calculates the air concentration resulting from the resuspension and dispersion of material located at the source and away from the source. These air concentrations are then integrated over time and area and multiplied by a population density to get the population inhalation exposure.

The model provides a basis for determining the importance of specific physical parameters and how changes in these parameters affect the population inhalation exposure. Its use is intended only to scope the magnitude of radiological impact on the surrounding population from a land area contaminated with radioactive material. The model is generic and is not directly applicable to a specific site because of the use of many simplifying assumptions.

Summary and Conclusions

A model is proposed for use in estimating the population inhalation exposure in the environs of an area contaminated with radioactive material. One use of this model is that the user can determine by sensitivity analysis, which parameters greatly affect the population inhalation exposure. Several simplifying assumptions were used in the model, such as uniform meteorology, uniform population density, point source approximation of an area source, constant deposition velocity, and constant resuspension rate and soil sink transfer rate. Also atmospheric dispersion versus distance is represented by a power function and a source depletion model is employed. These assumptions place strict limitations on the use of this model to specific sites.

To illustrate the use of the model, sample calculations were performed using selected parameter values. A comparison is made of two sets of parameters which differ only in the value of the resuspension rate, selected to represent typically observed initial and equilibrium rates. This, in effect, brackets the population inhalation exposure which would have been derived if the model used a time-dependent resuspension rate. The sample calculations use a 1 Curie Pu-239 source term as a reference level.

The following is a summary of the important results obtained for the sample calculation performed in this report. The use of parameter values, other than the ones used in this report, can result in different conclusions.

1. A high resuspension rate results in a greater population inhalation exposure than a low resuspension rate.

2. When a high resuspension rate exists, the population inhalation exposure is much greater from the secondary resuspension of contaminated material located away from the source, than the exposure from the initial resuspension of material located at the source.

3. For a low resuspension rate, the population inhalation exposure from the initial resuspension of contaminated material located at the source is much greater than the exposure from the secondary resuspension of material located away from the source.

4. For a high resuspension rate, most of the population inhalation exposure is delivered within the first year after a contaminating event, while for a low resuspension rate it takes 100 years for most of the exposure to be delivered.

5. The distance from the source within which most of the population inhalation exposure is delivered is dependent on deposition

velocity. For a doubling of the deposition velocity the distance within which most of the exposure occurs decreases by more than sixty-five per cent.

General Model Description

Figure 1 depicts the events considered in this model. The source is assumed to consist of a single radionuclide with an initial activity of 1 Curie. The contaminating event is assumed to occur as a ground level release, thus there is no initial airborne plume of material to consider. Contaminated material at the source is resuspended and dispersed resulting in an air concentration, X_0. Some fraction of this airborne contamination deposits on the ground, which also resuspends resulting in an air concentration X_1. Material from air concentration X_1 partially deposits on the ground, and resuspends resulting in air concentration X_2 and so on. Resuspension of contaminated material at the source will be called initial resuspension. Resuspension of material located outside the original source will be called secondary resuspension.

The model calculates, as a function of time and distance from the source, air concentrations, X_0 and X_1, and the respective population inhalation exposures, PE_0 and PE_1. The total population inhalation exposure, ΣPE_n, due to the sum of air concentrations, ΣX_n, is also calculated.

For sources consisting of several radionuclides, the population inhalation exposure is calculated separately for each radionuclide and multiplied by its activity and summed.

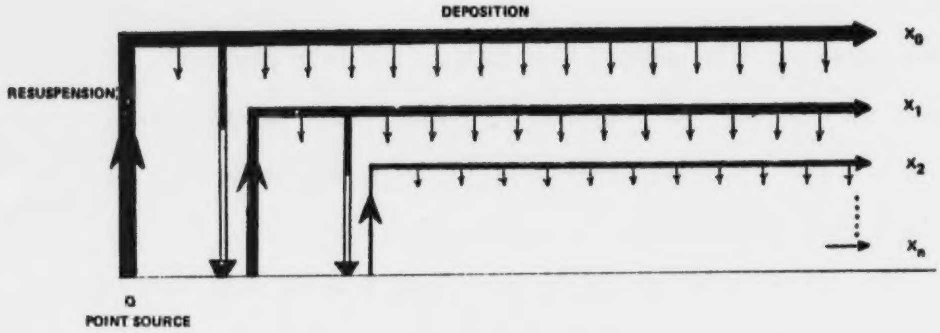


FIGURE 1

where:

Q = activity of point source (Ci)

r = distance from point source to receptor (m)

X_0 = air concentration at distance r and time t resulting from dispersion of resuspended material from source $(\frac{Ci}{m^3})$

X_1 = air concentration at distance r and time t resulting resuspension of material on ground at r $(\frac{Ci}{m^3})$

X_n = air concentration at distance r and time t resulting from resuspension of material deposited on ground from X_{n-1} $(\frac{Ci}{m^3})$

$X_T = \Sigma X_n$ = the air concentration at distance r and time t due to sum of initial and secondary resuspension $(\frac{Ci}{m^3})$

PE_0 = population inhalation exposure due to air concentration X_0(person - $\frac{Ci}{m^3}$ - sec)

PE_1 = population inhalation exposure due to air concentration X_1(person - $\frac{Ci}{m^3}$ - sec)

PE_n = population inhalation exposure due to air concentration X_n(person - $\frac{Ci}{m^3}$ - sec)

$PE_T = \Sigma PE_n$ = total population inhalation exposure (person - $\frac{Ci}{m^3}$ - sec)

Resuspension of contaminated material is the only physical process used to describe the movement of contaminated material from the soil to the air. Selected resuspension rates are assumed to be constant with time and distance.

The model was prompted by the recent concern about transuranium element soil contamination around several nuclear facilities in the United States. In general, only relatively small areas are involved, with population centers being fairly distant from the contamination (>1 km). Where larger areas were intially contaminated, the population centers are much farther away. Since the model's main purpose is to assess the total radiological impact over large areas, the area source is approximated by a point source. Where there is a small or no population near the source the point source approximation is satisfactory, considering the overall accuracy of the model.

Dispersion Equation

The air concentration of a radionuclide at a distance r from a continuous ground level point source is found by calculating the atmospheric dilution factor using a Gaussian diffusion equation which is a function of wind speed, Pasquill stability category (determines horizontal and vertical diffusion coefficients), and elevation of receptor.

If the source rate term, $\dot{Q}$, is known, the air concentration is:

$$X = \left(\frac{X}{\dot{Q}}\right)\dot{Q} \qquad (1)$$

where X = air concentration at distance r (Ci/m^3)

$X/\dot{Q}$ = atmospheric dilution factor at distance r (s/m^3)

$\dot{Q}$ = source rate (Ci/s)

Another method of determining the air concentration downwind from a source is from an equation for $X/\dot{Q}$ which is only a function of distance. The former Atomic Energy Commission has presented data on atmospheric dilution factors versus distance for a ground-level release for 17 nuclear power sites situated near rivers, lakes, and seacoasts. An equation which relates $X/\dot{Q}$ as a function distance can be empirically derived from this data (see Appendix I). This relationship is:

$$\frac{X}{\dot{Q}}(r) = \left(\frac{X}{\dot{Q}}\right)_{r_n} \left(\frac{r}{r_n}\right)^{-\ell} \qquad (2)$$

where r_n = normalizing distance (m)

$\left(\frac{X}{\dot{Q}}\right)_{r_n}$ = value of $X/\dot{Q}$ at $r = r_n$ (s/m^3)

$-\ell$ = slope of $X/\dot{Q}$ vs r on log-log plot

r = distance from source to receptor (m)

Equation (2) is valid only for those distances where data exist for $X/\dot{Q}$. It also not account for depletion of material as plume is dispersed. The deposition of material on the ground can be accounted for in two ways, either by considering depletion of material from the plume as it is dispersed, or by depletion of the source rate term $\dot{Q}$. Both methods yield the same result. A depletion correction factor based on depletion of the source term is (see Appendix I):

$$\frac{\dot{Q}^d}{\dot{Q}} = \exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right] \qquad (3)$$

where $\dot{Q}^d/\dot{Q}$ = depletion correction factor

$\dot{Q}^d$ = depleted source rate (Ci/s)

r_d = depletion distance = $\left[\frac{(2-\ell)r_n^{-\ell}}{2\pi V_d (X/\dot{Q})_{r_n}}\right]^{\frac{1}{2-\ell}}$ (m)

V_d = surface specific deposition velocity (m/s)

Equation (2) can be modified to account for deposition of material on the ground as follows:

$$\frac{X^d}{\dot{Q}^d}(r) = \left(\frac{X}{\dot{Q}}\right)_{r_n} \left(\frac{r}{r_n}\right)^{-\ell} \exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right] \qquad (4)$$

where $\frac{X^d}{\dot{Q}^d}(r) = \frac{X}{\dot{Q}}(r)$ corrected for ground deposition

In equation (3) the term r_d represents a depletion distance. When $r = r_d$, $\dot{Q}^d/\dot{Q}$ has decreased by a factor of 1/e or 37%.

This model assumes that equation (4) is applicable to all sites.

Calculation of Air Concentration Resulting from Resuspension of Material at Source and Subsequent Population Inhalation Exposure

Three loss mechanisms involved with the depletion of the point source activity are considered in this model. These are: resuspension of contamination from the ground, transfer of contaminated material from the soil surface to a soil sink, and radiological decay. Each loss mechanism is represented as an exponential decay with a specific decay constant and each is assumed to be independent of time. Thus the variation of the point source activity with time is:

$$Q(t) = Q \exp(-\lambda_t t) \qquad (5)$$

where $Q(t)$ = activity of point source at time t (Ci)

Q = initial activity of point source (Ci)

t = time (s)

λ_t = total decay constant = $\lambda_r + \lambda_s + \lambda_d$ (s^{-1})

λ_r = resuspension rate (s^{-1})

λ_s = transfer rate of contaminated material from soil surface to soil sink (s^{-1})

λ_d = radiological decay constant (s^{-1})

Equation (5) assumes there is no addition of contaminated material to point source thru time. The quantity of the source activity, Q(t), which may potentially result in an inhalation exposure to an individual is that amount being resuspended, which is:

$$\begin{aligned}\dot{Q}(t) &= \lambda_r Q(t)\\ &= \lambda_r Q \exp(-\lambda_t t)\end{aligned} \qquad (6)$$

where $\dot{Q}(t)$ = rate at which the source activity is being resuspended and available for dispersion (Ci/s)

The air concentration, X_0^d, of a radionuclide at a distance r from the source and at time t is the product of the atmospheric dilution factor and the source emission rate, that is:

$$X_0^d(r,t) = \frac{X^d}{\dot{Q}^d}(r) \cdot \dot{Q}(t) \qquad (7)$$

Substituting equations (4) and (6) into equation (7):

$$X_0^d(r,t) = \lambda_r Q \left(\frac{X}{\dot{Q}}\right)_{r_n} \left(\frac{r}{r_n}\right)^{-\ell} \exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right] \exp[-\lambda_t t] \qquad (8)$$

where X_0^d (r,t) = the air concentration of a radionuclide at a distance r from the source and at time t taking into account depletion of the plume. The subscript 0 denotes the air concentration

due to dispersion of contaminated material
from the point source.

Using polar coordinates, (r,Θ), the differential population inhalation exposure, dPE_0, in area dA at a radial distance r + dr from the point source and at an angle from a reference radial line is the product of the air concentration X_0^d and population density ρ in area dA during the time dt.

$$dPE_0 = X_0^d \rho dtdA \tag{9}$$

The total population inhalation exposure in area A and at time t is obtained by integrating equation (9) over area and time.

$$PE_0 = \int^A \int^t X_0^d \rho dtdA \tag{10}$$

where

$dA = rdrd\Theta$ (see Fig. 2)

ρ = population density

PE_0 = population inhalation exposure due to contaminated material resuspended from the source

From Fig. 2, $dA = rdrd\Theta$, substituting into equation (10)

$$PE_0(r,t) = \int_0^{2\pi} \int_{t_1}^{t_2} \int_{r_1}^{r_2} \rho X_0^d rdrdtd\Theta \tag{11}$$

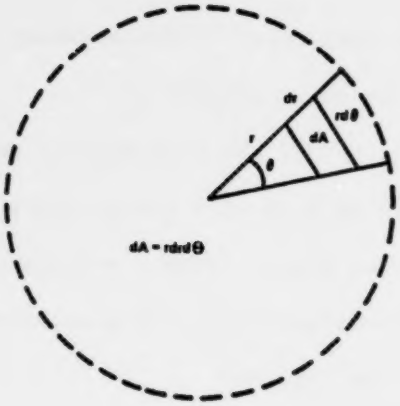


FIGURE 2

Assuming symmetry about the point source and uniform population density:

$$PE_0(r,t) = 2\pi\rho \int_{t_1}^{t_2} \int_{r_1}^{r_2} X_0^d \, rdrdt \qquad (12)$$

Substituting equation (8) for X_0^d into equation (12)

$$PE_0(r,t) = 2\pi\rho\lambda_r Q\left(\frac{X}{Q}\right)_{r_n} \int_{t_1}^{t_2} \int_{r_1}^{r_2} r\left(\frac{r}{r_n}\right)^{-\ell} \exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right] \cdot$$

$$\exp[-\lambda_t t]drdt \qquad (13)$$

Integrating over time and distance and simplifying gives: (see Appendix II for complete integration steps):

$$PE_0(r,t) = \frac{\rho Q}{V_d}\left(\frac{\lambda_r}{\lambda_t}\right)\left[\exp\left[-\left(\frac{r_1}{r_d}\right)^{2-\ell}\right] - \exp\left[-\left(\frac{r_2}{r_d}\right)^{2-\ell}\right]\right] \cdot \quad (14)$$

$$\left[\exp(-\lambda_t t_1) - \exp(-\lambda_t t_2)\right]$$

Evaluating equation (13) over the following limits, $r_1 = 0$, $r_2 = \infty$ and $t_1 = 0$, $t_2 = \infty$, gives:

$$PE_0(\infty,\infty) = \frac{\rho Q}{V_d}\left(\frac{\lambda_r}{\lambda_t}\right) \quad (15)$$

$PE_0(\infty,\infty)$ is referred to as the infinite population inhalation exposure. Integration over infinity is performed for mathematical convenience and represents a plateau for the population inhalation exposure. The plateau exposure is actually reached in a relatively short time and distance depending on the values of parameters used. Equation (14) can be evaluated over various times and distances to determine when and where the plateau exposure is achieved.

Discussion on the Derivation of the Air Concentrations X_1^d, X_2^d, ... X_n^d, Resulting from the Secondary Resuspension of Contaminated Material

Calculation of the air concentrations, X_1^d, X_2^d .... X_n^d, due to the secondary resuspension of contaminated material requires a prior determination of a soil surface concentration equation. The material in X_0^d deposits at a rate equal to $\omega_1 = V_d X_0^d$, where V_d is the surface specific deposition velocity. This deposited material results in a soil surface concentration denoted as Ω_1. The material that is a part of Ω_1 resuspends to give an air concentration X_1^d. The material X_1^d, as it is dispersed, deposits on the ground at a rate equal to $\omega_2 = V_d X_1^d$. This deposited material is a source term which results in a soil surface concentration denoted as Ω_2. The material that is a part of Ω_2 resuspends to give an air concentration X_2^d. The air concentrations X_3^d .... X_n^d are similarly derived. The total soil surface concentration at any point r from the original source is equal to the sum of the soil surface concentrations, Ω_1, Ω_2 .... Ω_n. Note that Ω_n is decreased by radiological decay and transfer to a soil sink. The actual air concentration at any point r is equal to the sum of air concentrations, X_0^d, X_1^d .... X_n^d. This model does not calculate the actual soil and air concentrations at any point r. Only the soil surface concentration Ω_1, and the air concentrations X_0^d and X_1^d are calculated. The result for the air concentration X_2^d is shown but is not calculated in this report. The population inhalation exposures PE_0, resulting from X_0, PE_1, resulting from X_1, and $PE_T(\infty,\infty)$ resulting from ΣX_n are calculated.

Calculation of Air Concentration X_1^d Resulting from the First Secondary Resuspension of Contaminated Material

$X_1^d(r,t)$ represents the air concentration at distance r and time t resulting from the first secondary resuspension of contaminated material. The material which is resuspended is that deposited from air concentration X_0^d. This deposited material is represented by the soil surface concentration Ω_1. Determining X_1^d requires calculation of the soil surface concentration, Ω_1, as a function of time and distance.

The soil surface concentration, Ω_1, at distance r and time t is increased by the deposition of contaminated material from X_0^d; it is decreased by resuspension of the deposited contamination, by transfer of the contaminated material from the soil surface to a soil sink, and by radiological decay. The importance of each loss mechanism depends largely on the value of its decay constant.

The deposition rate of contaminated material from air concentration X_0^d to the soil surface is:

$$\omega_1(r,t) = V_d X_0^d(r,t) \qquad \left(\frac{Ci}{m^2\text{-}s}\right) \tag{16}$$

The rate of change with time and distance of the soil surface concentration, Ω_1, can be expressed as:

$$\dot{\Omega}_1(r,t) = \underbrace{-\lambda_t\Omega_1(r,t)}_{\substack{\text{decrease in soil}\\ \text{surface concentration}}} + \underbrace{\omega_1(r,t)}_{\substack{\text{increase in soil}\\ \text{surface concentration}}} \qquad (17)$$

The total decay constant, λ_t, is equal to $\lambda_r + \lambda_s + \lambda_d$.

Since we are interested in the air concentration which results from a soil concentration at a distance r, equation (17) is changed from a partial differential equation to an ordinary differential equation by holding r constant and solving equation (17) with respect to time. The solution may be found in Appendix II. The result is:

$$\Omega_1(r,t) = V_d\lambda_r Q\left(\frac{X}{\dot{Q}}\right)_{r_n}\left(\frac{r}{r_n}\right)^{-\ell} \exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right] t\exp[-\lambda_t t] \qquad (18)$$

The time when the maximum soil surface concentration is reached can be calculated by setting the first time derivative of equation (18) equal to zero and solving for time t.

$$\frac{d\Omega_1(r,t)}{dt} = V_d\lambda_r Q\left(\frac{X}{\dot{Q}}\right)_{r_n}\left(\frac{r}{r_n}\right)^{-\ell} \exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right][\exp(-\lambda_t t)-\lambda_t t\exp(-\lambda_t t)]$$

$$\text{Let } A = V_d\lambda_r Q\left(\frac{X}{\dot{Q}}\right)_{r_n}\left(\frac{r}{r_n}\right)^{-\ell} \exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right]$$

then:

$$\frac{d\Omega_1(r,t)}{dt} = [A-A\lambda_t t][\exp(-\lambda_t t)] \qquad (19)$$

Setting equation (19) equal to zero and solving for t, the maximum soil surface concentration, Ω_1, is reached when:

$$t_{max} = \frac{1}{\lambda_t} \qquad (20)$$

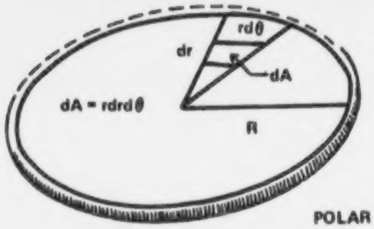
Now that the soil surface concentration as a function of time and distance is known, the air concentration, X_1^d can be calculated.

To calculate $X_1^d(r,t)$, the source rate, which is equal to $\lambda_r \Omega_1 A$, is multiplied by the depleted atmospheric dilution equation $X^d/\dot{Q}^d$ and integrated over area and time. In general terms this is:

$$X_1^d(r,t) = \int^{A} \int^{t} \lambda_r A \Omega_1 \left(\frac{X^d}{\dot{Q}^d}\right) dA dt \qquad (21)$$

Equation (4) shows that $X^d/\dot{Q}^d$ is a function of distance r and equation (18) shows that Ω_1 is a function of distance r. When these terms are multiplied the result becomes very difficult to integrate explicitly. The following section presents a method by which the integration of $X_1^d(r,t)$ becomes simplified.

Consider a disk of radius R uniformly contaminated with a soil surface concentration Ω_1 (Ci/m^2) located at a distance r from the original source. See Figure 3. The source rate $\dot{Q}$ from this disk is the product of the soil concentration, Ω_1, resuspension rate, λ_r,



POLAR COORDINATES

FIGURE 3

and the area, A, of the disk. The incremental source rate in area dA is:

$$d\dot{Q} = \Omega_1 \lambda_r r dr d\Theta \qquad (22)$$

The air concentration, X_1^d, at the center of this disk is the area integral of the source rate times the depleted atmospheric dilution factor, X^d/Q^d, given in equation (4). Remember Ω_1 is now assumed to be constant.

$$X_1^d = \int_0^{2\pi} \int_0^R \lambda_r \Omega_1 \left(\frac{X}{\dot{Q}}\right)_{r_n} \left(\frac{r}{r_n}\right)^{-\ell} \exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right] r dr d\Theta \qquad (23)$$

Symmetry is assumed, thus the integral for dΘ is equal to 2π. Integration over distance r was performed earlier in a similar calculation and the solution may be found in Appendix II, part A. The result is:

$$X_1^d = \frac{\lambda_r \Omega_1}{V_d} [1 - \exp(-\frac{R}{r_d})^{2-\ell}] \qquad (24)$$

For a disk radius $R > r_d$, the depletion distance, the term $\exp[-(\frac{R}{r_d})^{2-\ell}]$ approaches zero, thus the air concentration becomes:

$$X_1^d = \frac{\lambda_r \Omega_1}{V_d} \qquad (25)$$

Equation (25) gives the air concentration at the center of a disc which is uniformly contaminated. Equation (18) gives the variation of the soil surface concentration Ω_1 with time and distance., Substituting equation (18) for Ω_1 in equation (25) gives:

$$X_1^d(r,t) = \lambda_r^2 \, Q (\frac{X}{\dot{Q}})_{r_n} (\frac{r}{r_n})^{-\ell} \exp[-(\frac{r}{r_d})^{2-\ell}] \, t \exp[-\lambda_t t] \qquad (26)$$

The validity of the uniformly contaminated disk assumption may be examined by looking at $\Omega_1(r,t)$ plotted as a function of distance (equation 18). The soil surface concentration, $\Omega_1(r,t)$ is an exponential function of distance r. This says that for points close to the source the soil surface concentration drops rapidly with distance and for points far from the source $\Omega_1(r,t)$ decreases less rapidly. The uniformly contaminated disk assumption becomes more valid the further one gets from the source. The model is mainly interested in the total population inhalation exposure and not so much with the variation of exposure with distance from the source. Any overestimate and underestimate with distance of the air concentration is averaged out because of the uniform population density assumption.

Calculation of the Population Inhalation Exposure, $PE_1(r,t)$ Resulting from Air Concentration $X_1^d(r,t)$

The population inhalation exposure, PE_1, due to air concentration X_1^d is derived the same way PE_0 was calculated. That is:

$$PE_1(r,t) = \int_0^{2\pi} \int_{t_1}^{t_2} \int_{r_1}^{r_2} \rho X_1^d r\,dr\,dt\,d\Theta \qquad (27)$$

Assuming symmetry and uniform population density,

$$PE_1(r,t) = 2\pi\rho \int_{t_1}^{t_2} \cdot \int_{r_1}^{r_2} X_1^d r\,dr\,dt \qquad (28)$$

Substituting equation (26) for X_1^d in equation (28);

$$PE_1(r,t) = 2\pi\rho\lambda_r^2 \left(\frac{X}{\dot{Q}}\right)_{r_n} \int_{t_1}^{t_2} \int_{r_1}^{r_2} r\left(\frac{r}{r_d}\right)^{-\ell} \exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right] \cdot \qquad (29)$$

$$t\exp[-\lambda_t t]\,dr\,dt$$

Integrating over time and distance and simplifying gives: (see Appendix II for complete integration steps).

$$PE_1(r,t) = \frac{\rho Q \lambda_r^2}{V_d}\left[\exp\left[-\left(\frac{r_1}{r_d}\right)^{2-\ell}\right]-\exp\left[-\left(\frac{r_2}{r_d}\right)^{2-\ell}\right]\right] \cdot \left[\left(\frac{\lambda_t t_1 + 1}{\lambda_t^2}\right)\exp(-\lambda_t t_1) - \left(\frac{\lambda_t t_2 + 1}{\lambda_t^2}\right)\exp(-\lambda_t t_2)\right] \qquad (30)$$

Evaluating equation (30) for $r_1 = 0$, $r_2 = \infty$ and $t_1 = 0$, $t_2 = \infty$ gives:

$$PE_1(\infty,\infty) = \frac{\rho Q}{V_d}\left(\frac{\lambda_r}{\lambda_t}\right)^2 \qquad (31)$$

Calculation of the Total Infinite Population Inhalation Exposure $PE_T(\infty,\infty)$

Previous sections have calculated the population inhalation exposures due to the initial resuspension of material at the point source and from the first secondary resuspension of material at distance r that originated from the point source, i.e., PE_0 and PE_1. The material at distance r which deposited from air concentration X_1^d is also available for resuspension and results in air concentration X_2^d. The material associated with X_2^d is dispersed and deposited, and resuspends as X_3^d and so on. For each of the air concentrations X_n^d, there is an associated population inhalation exposure, PE_n. The summation of all the population inhalation exposures, $\Sigma\, PE_n$, gives the total population inhalation exposure, PE_T.

A simple expression for the total infinite population inhalation exposure, $PE_T\ (\infty,\infty)$ can be derived. Equations (16) and (31) for $PE_0\ (\infty,\infty)$ and $PE_1\ (\infty,\infty)$ respectively show that $PE_1(\infty,\infty)$ differs from $PE_0\ (\infty,\infty)$ by a factor of $\frac{\lambda_r}{\lambda_t}$, i.e.:

$$PE_0(\infty,\infty) = \frac{\rho Q}{V_d}\left(\frac{\lambda_r}{\lambda_t}\right) \qquad PE_1(\infty,\infty) = \frac{\rho Q}{V_d}\left(\frac{\lambda_r}{\lambda_t}\right)^2$$

The air concentration X_2^d, is derived by the same method as X_1^d was determined except Ω_1 becomes Ω_2 and the deposition rate becomes $\omega_2 = V_d X_1^d$. The infinite population inhalation exposure, $PE_2(\infty,\infty)$ is the integral of the air concentration $X_2^d(r,t)$ multiplied by the population density. The result is:

$$PE_2(\infty,\infty) = \frac{\rho Q}{V_d}\left(\frac{\lambda_r}{\lambda_t}\right)^3 \qquad (32)$$

Each air concentration, X_n^d, results in an infinite population inhalation exposure, PE_n, which differs from the previous exposure, PE_{n-1}, by a factor of $\frac{\lambda_r}{\lambda_t}$. Therefore the total infinite population inhalation exposure is:

$$PE_T(\infty,\infty) = PE_0(\infty,\infty) + PE_1(\infty,\infty) + PE_2(\infty,\infty) + \dots + PE_n(\infty,\infty)$$

$$PE_T(\infty,\infty) = \frac{\rho Q}{V_d}\left(\frac{\lambda_r}{\lambda_t}\right) + \frac{\rho Q}{V_d}\left(\frac{\lambda_r}{\lambda_t}\right)^2 + \frac{\rho Q}{V_d}\left(\frac{\lambda_r}{\lambda_t}\right)^3 + \dots + \frac{\rho Q}{V_d}\left(\frac{\lambda_r}{\lambda_t}\right)^n$$

$$PE_T(\infty,\infty) = \sum_{j=1}^{n} \frac{\rho Q}{V_d}\left(\frac{\lambda_r}{\lambda_t}\right)^j \qquad (33)$$

Equation (33) is a geometric series, which stated in a more general form is:

$$\sum_{j=m}^{n} ar^j = \frac{ar^m(1-r^{n-1+m})}{1-r} \qquad (34)$$

When $r < 1$ and $m = 1$ and $n = \infty$, equation (34) reduces to:

$$\sum_{j=1}^{\infty} ar^j = \frac{ar}{1-r} \qquad (35)$$

In equation (33), $a = \frac{\rho Q}{V_d}$ and $r = \frac{\lambda_r}{\lambda_t}$ which is always less than one, therefore:

$$PE_T(\infty,\infty) = \frac{\frac{\rho Q}{V_d}\left(\frac{\lambda_r}{\lambda_t}\right)}{1-\frac{\lambda_r}{\lambda_t}}$$

Substituting in $\lambda_t = \lambda_r + \lambda_s + \lambda_d$ and simplifying gives:

$$PE_T(\infty,\infty) = \frac{\rho Q}{V_d}\left(\frac{\lambda_r}{\lambda_d+\lambda_s}\right) \qquad (36)$$

Environmental Dose Commitment

Environmental dose commitment is defined as "the sum of all doses to individuals over the entire time period the material persists in the environment in a state available for interaction with humans" and has units of person-rem. "It is calculated for a specific release at a specific time and is obtained by summing the person-rem delivered in each of the years following release to the environment until dose increments are inconsequential by other means" (1).

The environmental dose commitment resulting from the inhalation pathway in this model is calculated by multiplying the total infinite population inhalation exposure by an annual breathing rate and a dose conversion factor, i.e.:

$$\text{Environmental Dose Commitment (person-rem)} = \qquad (37)$$

$$\text{Total Infinite Population Inhalation Exposure } \left(\text{person} - \frac{\text{Ci}}{\text{m}^3} - \text{yr}\right) \times$$

$$\text{Breathing Rate } \left(\frac{\text{m}^3}{\text{y}}\right) \times \text{Dose Conversion Factor } \left(\frac{\text{rem}}{\text{Ci}}\right)$$

The total infinite population inhalation exposure, designated as $PE_T(\omega,\infty)$, is directly obtained by integrating over all distance and time, the air concentration $X_n^d(r,t)$ multiplied by the population density, ρ. The annual breathing rate gives the total volume of air inhaled by a person in one year. The dose conversion factor converts the activity of a radionuclide inhaled into a dose, and is dependent on many parameters, such as organ of interest, radionuclide inhaled, activity mean aerodynamic diameter (AMAD) of particles inhaled, and biological residence time in organ.

The environmental dose commitment can also be calculated for a period of time less than that stated in equation (37). For example, a 100 year environmental dose commitment is determined by calculating the population inhalation exposure committed over a 100 year period and substituting this value for the total infinite population inhalation exposure in equation (37).

The environmental dose commitment may be used to calculate the cumulative potential health effects that result from a release of radioactive material into the environment by multiplying the environmental dose commitment for each organ by a dose-risk conversion factor. Dose-risk conversion factors are derived for somatic and genetic risk, and are dependent on the organ of interest, radionuclide, and age category of person, usually child or adult.

The following sample calculation illustrates the conversion of population inhalation exposure to an environmental dose commitment. The calculation will determine the environmental dose commitment for the pulmonary region of the lung as the result of inhaling Pu-239 particles. The pulmonary region of the lung is considered the organ at greatest risk for the inhalation of Pu-239 particles.

Equation (37) requires data for 3 parameters to calculate the environmental dose commitment, the total infinite population inhalation exposure, the annual breathing rate of a person, and a dose conversion factor. From Table 2, the total infinite population inhalation exposure for parameter set #1 (see Table 1), is 8.5×10^{-11} person-Ci yr/m^3. The annual breathing rate is assumed to be that of standard man and is equal to 8395 m^3/yr (2).

The dose conversion factor (DCF) is calculated using the following equation:

$$DCF = 7.38\times10^{-2}\,\frac{ET_E fa}{m}\,\frac{\text{mrad}}{\text{pCi inhaled}} \qquad (38)$$

where:

E = effective energy of disintegrations MeV/dis

T_E = effective half-life of radionuclide in organ days

f_a = fraction of inhaled particles reaching organ

m = mass of organ grams

The effective energy of Pu-239 disintegrations is 5.15 MeV/dis (3). The effective half-life of Pu-239 in the lung is equal to the biological half-life since the radiological half-life of Pu-239 is so long. Thus the effective half-life of Pu-239 in the lung is 300.4 days assuming the Pu-239 particles to be a class Y compound (4). The fraction of Pu-239 particles inhaled reaching the pulmonary region of lung is .23, assuming the Pu-239 particles have an AMAD of 1 micron (5). The mass of the pulmonary region of the lung is 570 grams (2). Substituting these parameter values into equation (38) gives:

$$DCF = 7.38\times10^{-2}\,\frac{(5.15)(300.4)(.23)}{570}\,\frac{\text{mrad}}{\text{pCi inhaled}}$$

$$DCF = .046 \frac{\text{mrad}}{\text{pCi inhaled}} = 4.6\text{x}10^{7} \frac{\text{rad}}{\text{Ci inhaled}}$$

If a quality factor of 20 is assumed (9), the dose conversion factor becomes $9.2\text{x}10^{8}$ rem/Ci inhaled. From equation (37), the environmental dose commitment is:

$$EDC = (8.5\text{x}10^{-11} \text{ person} - \frac{\text{Ci}}{\text{m}^3} - \text{yr})(8395 \frac{\text{m}^3}{\text{yr}})(9.3\text{x}10^{8} \frac{\text{rem}}{\text{Ci inhaled}})$$

$$EDC = 656 \text{ person-rem}$$

Equation (38) is applicable only for an acute intake of radioactive material. This calculation assumed that the exposure, $PE_T(\infty,\infty)$, occurs as the of an acute intake. Figure 5 shows for parameter set #1 that $PE_T(\infty,\infty)$ is reached within one year. If the total infinite population inhalation exposure is committed over a period of greater than one year, the acute intake assumption is not valid and the dose conversion factor must be derived for a continous intake situation.

The use of the phrase "infinite dose" is used in the same context as "infinite exposure" whose meaning is explained on page (13).

Parameter Selection

Table 1 presents the values of parameters selected to illustrate exposure calculations using the model. Parameter values selected are

Table 1

Parameter Values Used To Illustrate Model

Set #1	Set #2
$\lambda_r = 10^{-7}\ s^{-1}$	$\lambda_r = 10^{-11}\ s^{-1}$
$\lambda_t = 1.01\text{x}10^{-7}\ s^{-1}$	$\lambda_t = 1.01\text{x}10^{-9}\ s^{-1}$

$\lambda_s = 10^{-9}\ s^{-1}$

$\lambda_d = 10^{-12}\ s^{-1}$

$V_d = 0.01\ \frac{m}{s}$

$Q = 1\ Ci$

$r_n = 1000\ m$

$\left(\frac{X}{\dot{Q}}\right)_{1km} = 2.5\text{x}10^{-6}\ \frac{s}{m^3}$

$\ell = 1.43$

$r_d = 9595\ m$

$\rho = 2.7\text{x}10^{-5}\ \frac{\text{persons}}{m^2}$

intended to correspond to typically observed values in the field, but are not indicative of any particular site. Two calculations are made differing only in the value of the resuspension rate, λ_r, and therefore the total decay constant, λ_t. The high resuspension rate represents an area recently contaminated or an area being mechanically disturbed, while the low resuspension rate represents a typical aged source. Current data indicates that the resuspension rate is a function of time, but this model does not take into account the time dependence of the resuspension rate; it is assumed that the resuspension rate remains constant throughout time and also distance.

The model also depends on values chosen for the transfer rate of contamination from soil surface to the soil sink, population density, deposition velocity, half-life of the radionuclide of interest, and the source strength. The transfer rate from the soil surface to the soil sink was taken from the report ORNL-4992 (6), while the population density used is that of the continental United States during 1970. Deposition velocity is a complex parameter dependent on the meteorology of the site, and particle-soil surface aerodynamic properties (7). The value selected for the deposition velocity is one typically found in the field and is assumed to be constant with time and distance.

Since this model is addressed to transuranium element contamination, in particular Pu-239, radiological decay will generally not be of any consequence in calculating population inhalation exposure. The source strength is arbitrarily taken to be 1 Ci of Pu-239.

For source activities greater or less than 1 Ci and for different radionuclides, the population inhalation exposure calculated for each radionuclide are multiplied by their respective activities and summed to get the total population inhalation exposure. The specific parameter values of the dispersion equation used in this model are discussed further in Appendix I.

Results

Table 2 gives the population inhalation exposure, $PE(\infty,\infty)$, integrated over distance, $r = 0 \rightarrow \infty$, and time, $t = 0 \rightarrow \infty$. When the resuspension rate is high (10^{-7} s^{-1}), the exposure PE_1 from the first secondary resuspension is almost equal to the exposure, PE_0 from the initial resuspension. The sum of PE_0 and PE_1 is a factor of 51 lower than the total exposure indicating that subsequent resuspension after the first secondary resuspension is a significant contributor to the total infinite population inhalation exposure for the high resuspension rate.

For the low resuspension rate (10^{-11} s^{-1}) the exposure PE_0 from the initial resuspension is a factor of 100 higher than the exposure, PE_1, from the first secondary resuspension. The sum of PE_0 and PE_1 is just about equal to the total exposure indicating that secondary resuspension is not much of a contributor to the total infinite population inhalation exposure for the low resuspension rate.

The total infinite population inhalation exposure, $PE_T(\infty,\infty)$, is a factor of 10^4 higher for the resuspension rate of 10^{-7} s^{-1} as compared to the resuspension rate of 10^{-11} s^{-1}, with all other parameters equal. If a time dependent resuspension rate was used in this model instead of a constant resuspension rate, the total infinite population inhalation exposure would be expected to lie between the exposures resulting from the resuspension rates of 10^{-7} s^{-1} and 10^{-11} s^{-1}, assuming these are the initial and final resuspension rates.

Table 3 presents the population inhalation exposure as a function of time integrated over the distance $r=0\rightarrow\infty$. The high resuspension rate results in most of the population inhalation exposure being delivered within the first year after a contaminating event, while for the low resuspension it takes 100 years after a contaminating event for most of the population inhalation exposure to be delivered. Figure 4 is a plot of the values in Table 3.

Table 4 gives the distance within which the fraction X of the total infinite population inhalation exposure is delivered for various deposition velocities. Even though the total infinite population inhalation exposure is obtained by integrating over infinite distance, 95% of the exposure is delivered, for example, within 66 kilometers for a deposition velocity of 0.01 m/s. Doubling the deposition velocity decreases the distance within most of the infinite population inhalation exposure occurs by more than sixty-five per cent.

Tables 5 and 6 presents the air concentration as a function of distance at various times after a contaminating event, taking into account depletion. For the high resuspension rate the air concentration, X_1^d, resulting from the first secondary resuspension is greater than the air concentration, X_0^d, resulting from the initial resuspension of the contaminated material. Within 10 years both air concentrations drop to very small levels. For the low resuspension rate the air concentration, X_0^d, resulting from the initial resuspension is much higher than the air concentration resulting from the first secondary resuspension. After 50 years both air concentrations are still within a factor of 10 of the levels occurring after one year. Figures 5 and 6 are a plot of the values in Tables 5 and 6.

Tables 7 and 8 gives air concentration as a function of time at various distances, taking into account depletion. For the high resuspension the air concentrations essentially drops to zero, while for the low resuspension rate the air concentrations drop only by a factor of 100 after 100 years.

Table 9 gives the soil concentration Ω_1 resulting from deposition of material originating from source as a function of distance, 1 year and 10 years after a contaminating event. The high resuspension rate has a 10 year soil concentration much lower than the 1 year soil concentration greater than the 1 year soil concentration. See Figures 7 and 8.

Tables 10 and 11 presents the soil concentration, Ω_1, as a function of time at various distances. The maximum soil concentration is reached in 0.31 years for the high resuspension rate and in 31.43 years for the low resuspension rate. See Figure 9.

Table 2

Infinite* Population Inhalation Exposure

	Parameter Set #1	Parameter Set #2
	(person - $\frac{Ci}{m^3}$ - y)	(person - $\frac{Ci}{m^3}$ - y)
$PE_0(\infty,\infty)$	8.5×10^{-11}	8.5×10^{-13}
$PE_1(\infty,\infty)$	8.4×10^{-11}	8.4×10^{-15}
$PE_T(\infty,\infty)$	8.6×10^{-9}	8.6×10^{-13}

* Refers to integration over all distance and time, 95% of exposure occurs within 66 km from source for both parameter sets and levels stated above reached in 1y and 100y for parameter sets 1 and 2 respectively.

Table 3

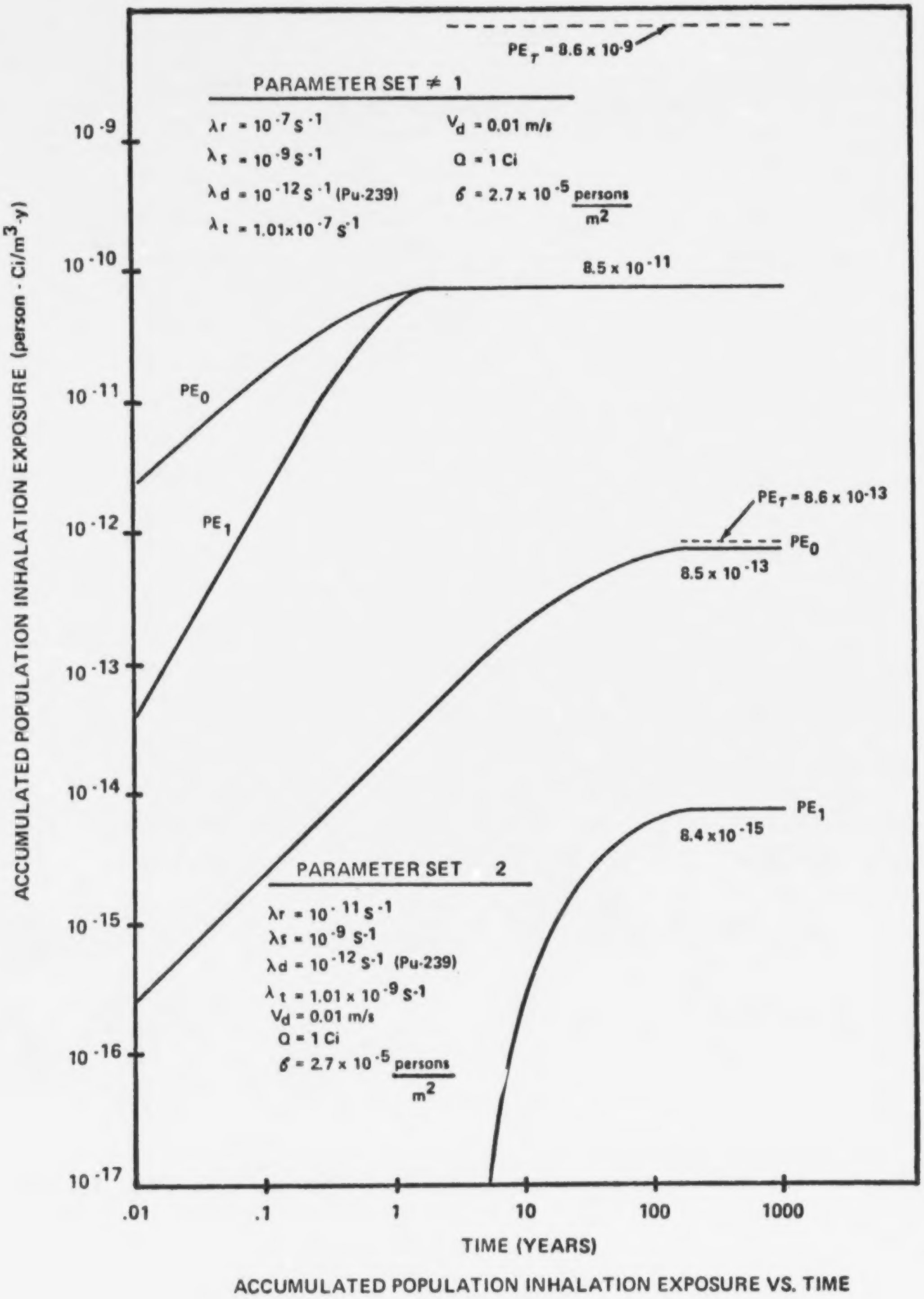
Accumulated Population Inhalation Exposure* As A Function Of Time

	Parameter Set #1		Parameter Set #2	
t_2 (y)	PE_0 (person - $\frac{Ci}{m^3}$ - y)	PE_1 (person - $\frac{Ci}{m^3}$ - y)	PE_0 (person - $\frac{Ci}{m^3}$ - y)	PE_1 (person - $\frac{Ci}{m^3}$ - y)
0.01	2.7×10^{-12}	4.2×10^{-14}	2.7×10^{-16}	4.3×10^{-22}
0.1	2.3×10^{-11}	3.4×10^{-12}	2.7×10^{-15}	4.2×10^{-20}
1	8.1×10^{-11}	6.9×10^{-11}	2.7×10^{-14}	4.2×10^{-18}
10	8.5×10^{-11}	8.4×10^{-11}	2.3×10^{-13}	3.4×10^{-16}
25	"	"	4.7×10^{-13}	1.6×10^{-15}
50	"	"	6.8×10^{-13}	4.0×10^{-15}
100	"	"	8.1×10^{-13}	6.9×10^{-15}
250	"	"	8.5×10^{-13}	8.4×10^{-15}
500	"	"	"	"
1000	"	"	"	"

$$PE_0(t) = \frac{\rho Q}{V_d}\left(\frac{\lambda_r}{\lambda_t}\right)[1-\exp(-\lambda_t t_2)]$$

$$PE_1(t) = \frac{\rho Q}{V_d}\left(\frac{\lambda_r}{\lambda_t}\right)^2 [1-(\lambda_t t_2+1)\exp(-\lambda_t t_2)]$$

*integrated over all distance.



ACCUMULATED POPULATION INHALATION EXPOSURE VS. TIME

FIGURE 4

Table 4

Distance Within Which The Fraction X of Accumulated Exposure Is Delivered As A Function Of V_d

	r_2 (m)		
X	$V_d=0.02\ \frac{m}{s}$*	$V_d=0.01\ \frac{m}{s}$**	$V_d=0.005\ \frac{m}{s}$***
.05	16	52	177
.10	55	185	625
.25	320	1,078	3,638
.35	649	2,190	7,387
.45	1,153	3,891	13,128
.50	1,495	5,044	17,017
.60	2,440	8,231	27,767
.75	5,044	17,018	57,413
.90	12,286	41,449	139,833
.92	14,450	48,751	164,469
.95	19,494	65,768	221,877
.98	31,134	105,038	354,359
.99	41,449	139,841	471,771
.999	89,421	284,817	960,868
.9999	139,843	471,799	1,591,675

$$r_2 = r_d[-\ell n(1-x)]^{\frac{1}{2-\ell}}$$

* r_d = 2,844m, ℓ=1.43 ** r_d = 9594m, ℓ=1.43 *** r_d = 32,370m, ℓ=1.43

Table 5

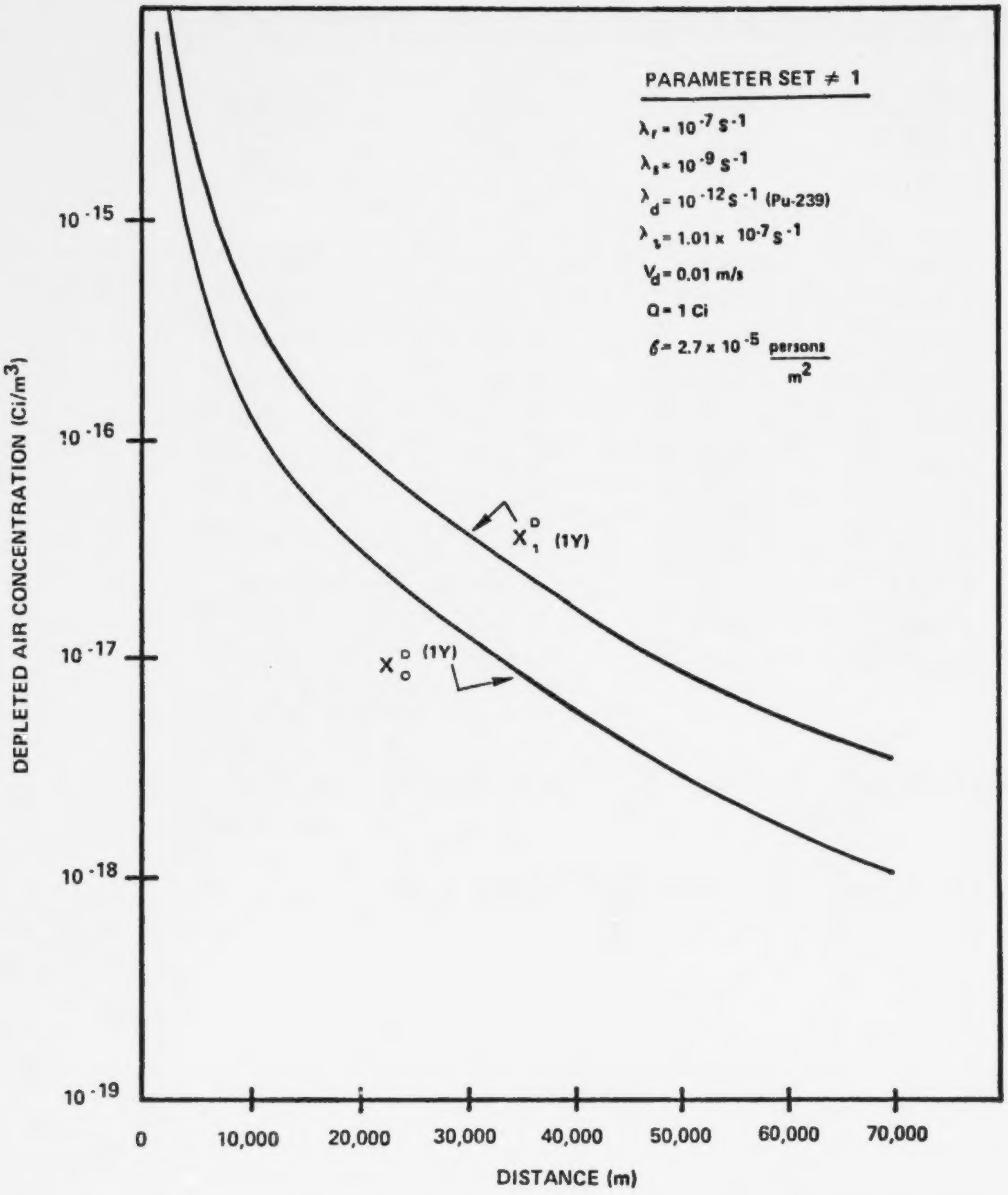
Air Concentration* As A Function Of Distance-Parameter Set #1

r (m)	$X_0^d(1y)$ $(\frac{Ci}{m^3})$	$X_0^d(10y)$ $(\frac{Ci}{m^3})$	$X_1^d(1y)$ $(\frac{Ci}{m^3})$	$X_1^d(10y)$ $(\frac{Ci}{m^3})$
1,000	7.8×10^{-15}	2.8×10^{-27}	2.5×10^{-14}	8.8×10^{-26}
10,000	1.4×10^{-16}	4.9×10^{-29}	4.4×10^{-16}	1.5×10^{-27}
20,000	3.1×10^{-17}	1.1×10^{-29}	9.8×10^{-17}	3.5×10^{-28}
30,000	1.2×10^{-17}	4.2×10^{-30}	3.7×10^{-17}	1.3×10^{-28}
40,000	5.5×10^{-18}	2.0×10^{-30}	1.7×10^{-17}	6.2×10^{-29}
50,000	3.0×10^{-18}	1.0×10^{-30}	9.4×10^{-18}	3.3×10^{-29}
60,000	1.7×10^{-18}	6.1×10^{-31}	5.4×10^{-18}	1.9×10^{-29}
70,000	1.1×10^{-18}	3.8×10^{-31}	3.4×10^{-18}	1.2×10^{-29}
80,000	6.9×10^{-19}	2.4×10^{-31}	2.2×10^{-18}	7.7×10^{-30}
90,000	4.6×10^{-19}	1.6×10^{-31}	1.5×10^{-18}	5.2×10^{-30}
100,000	3.2×10^{-19}	1.1×10^{-31}	1.0×10^{-18}	3.5×10^{-30}

$$X_0^d(r,t) = \lambda_r Q\left(\frac{X}{\dot{Q}}\right)_{r_n}\left(\frac{r}{r_n}\right)^{-\ell}\exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right]\exp[-\lambda_t t]$$

$$X_1^d(r,t) = \lambda_r^{2} Q\left(\frac{X}{\dot{Q}}\right)_{r_n}\left(\frac{r}{r_n}\right)^{-\ell}\exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right]t\ \exp[-\lambda_t t]$$

*corrected for deposition from plume



AIR CONCENTRATION VS. DISTANCE – PARAMETER SET ≠ 1

FIGURE 5

Table 6

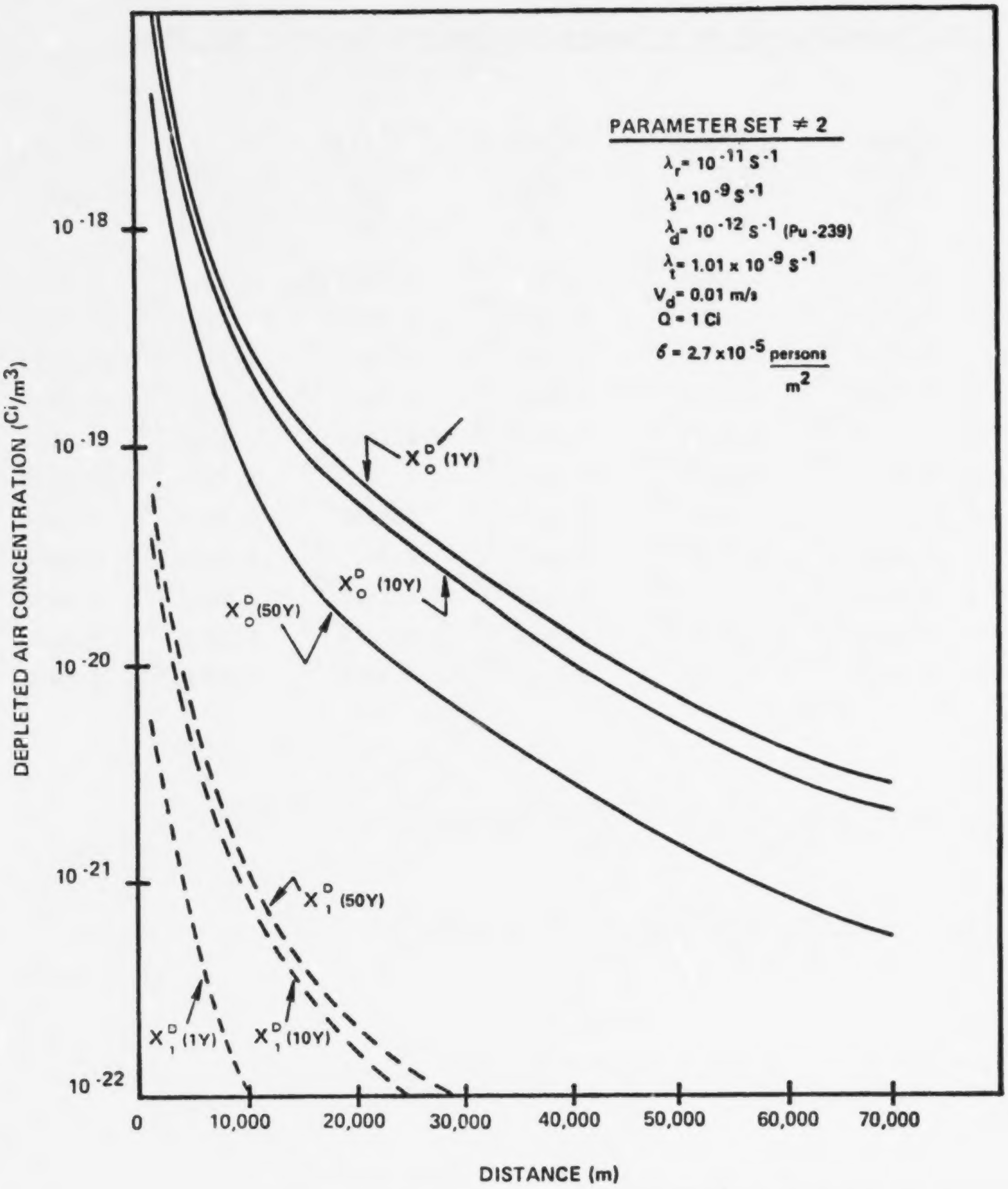
Air Concentration* As A Function Of Distance-Parameter Set #2

r (m)	$X_0^d(1y)$ $(\frac{Ci}{m^3})$	$X_0^d(10y)$ $(\frac{Ci}{m^3})$	$X_0^d(50y)$ $(\frac{Ci}{m^3})$	$X_1^d(1y)$ $(\frac{Ci}{m^3})$	$X_1^d(10y)$ $(\frac{Ci}{m^3})$	$X_1^d(50y)$ $(\frac{Ci}{m^3})$
1,000	$1.8\text{x}10^{-17}$	$1.4\text{x}10^{-17}$	$3.9\text{x}10^{-18}$	$5.8\text{x}10^{-21}$	$4.4\text{x}10^{-20}$	$6.1\text{x}10^{-20}$
10,000	$3.2\text{x}10^{-19}$	$2.4\text{x}10^{-19}$	$6.8\text{x}10^{-20}$	$1.0\text{x}10^{-22}$	$7.7\text{x}10^{-22}$	$1.1\text{x}10^{-21}$
20,000	$7.7\text{x}10^{-20}$	$5.5\text{x}10^{-20}$	$1.5\text{x}10^{-20}$	$2.3\text{x}10^{-23}$	$1.7\text{x}10^{-22}$	$2.4\text{x}10^{-22}$
30,000	$2.7\text{x}10^{-20}$	$2.1\text{x}10^{-20}$	$5.8\text{x}10^{-21}$	$8.7\text{x}10^{-24}$	$6.5\text{x}10^{-23}$	$9.1\text{x}10^{-23}$
40,000	$1.3\text{x}10^{-20}$	$9.5\text{x}10^{-21}$	$2.7\text{x}10^{-21}$	$4.1\text{x}10^{-24}$	$3.1\text{x}10^{-23}$	$9.3\text{x}10^{-23}$
50,000	$6.9\text{x}10^{-21}$	$5.2\text{x}10^{-21}$	$1.5\text{x}10^{-21}$	$2.2\text{x}10^{-24}$	$1.6\text{x}10^{-23}$	$2.3\text{x}10^{-23}$
60,000	$4.0\text{x}10^{-21}$	$3.0\text{x}10^{-21}$	$8.5\text{x}10^{-22}$	$1.3\text{x}10^{-24}$	$9.6\text{x}10^{-24}$	$1.3\text{x}10^{-23}$
70,000	$2.5\text{x}10^{-21}$	$1.9\text{x}10^{-21}$	$5.2\text{x}10^{-22}$	$7.9\text{x}10^{-25}$	$5.9\text{x}10^{-24}$	$8.3\text{x}10^{-24}$
80,000	$1.6\text{x}10^{-21}$	$1.2\text{x}10^{-21}$	$3.4\text{x}10^{-22}$	$5.1\text{x}10^{-25}$	$3.8\text{x}10^{-24}$	$5.3\text{x}10^{-24}$
90,000	$1.1\text{x}10^{-21}$	$8.1\text{x}10^{-22}$	$2.3\text{x}10^{-22}$	$3.4\text{x}10^{-25}$	$2.6\text{x}10^{-24}$	$3.6\text{x}10^{-24}$
100,000	$7.4\text{x}10^{-22}$	$5.6\text{x}10^{-22}$	$1.6\text{x}10^{-22}$	$2.3\text{x}10^{-25}$	$1.8\text{x}10^{-24}$	$2.5\text{x}10^{-24}$

$$X_0^d(r,t) = \lambda_r Q\left(\frac{X}{\dot{Q}}\right)_{r_n}\left(\frac{r}{r_n}\right)^{-\ell}\exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right]\exp[-\lambda_t t]$$

$$X_1^d(r,t) = \lambda_r^2 Q\left(\frac{X}{\dot{Q}}\right)_{r_n}\left(\frac{r}{r_n}\right)^{-\ell}\exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right]t\ \exp[-\lambda_t t]$$

*corrected for deposition from plume



AIR CONCENTRATION VS. DISTANCE – PARAMETER SET ≠ 2
FIGURE 6

Table 7

Air Concentration* As A Function Of Time At Various Distances-Parameter Set #1

t (y)	$X_0^d(\frac{Ci}{m^3})$			$X_1^d(\frac{Ci}{m^3})$		
	r=1,000m	r=10,000m	r=20,000m	r=1,000m	r=10,000m	r=20,000m
0.01	$1.8x10^{-13}$	$3.2x10^{-15}$	$7.3x10^{-16}$	$5.8x10^{-15}$	$1.0x10^{-16}$	$2.3x10^{-17}$
0.1	$1.4x10^{-13}$	$2.4x10^{-15}$	$5.5x10^{-16}$	$4.4x10^{-14}$	$7.7x10^{-16}$	$1.7x10^{-16}$
1	$7.8x10^{-15}$	$1.4x10^{-16}$	$3.1x10^{-17}$	$2.5x10^{-14}$	$4.4x10^{-16}$	$9.8x10^{-17}$
10	$2.8x10^{-27}$	$4.9x10^{-29}$	$1.1x10^{-29}$	$8.8x10^{-26}$	$1.5x10^{-27}$	$3.5x10^{-28}$
25	$4.9x10^{-48}$	$8.6x10^{-50}$	$2.0x10^{-50}$	$3.9x10^{-46}$	$6.8x10^{-48}$	$1.5x10^{-48}$
50	$<10^{-50}$	$<10^{-50}$	$<10^{-50}$	$<10^{-50}$	$<10^{-50}$	$<10^{-50}$
100	"	"	"	"	"	"
250	"	"	"	"	"	"
500	"	"	"	"	"	"
1000	"	"	"	"	"	"

*corrected for deposition from plume

Table 8

Air Concentration* As A Function Of Time At Various Distances-Parameter Set #2

$\frac{t}{(y)}$	$X_0^d(\frac{Ci}{m^3})$			$X_1^d(\frac{Ci}{m^3})$		
	r=1,000m	r=10,000m	r=20,000m	r=1,000m	r=10,000m	r=20,000m
0.01	1.9×10^{-17}	3.3×10^{-19}	7.5×10^{-20}	6.0×10^{-23}	1.1×10^{-24}	2.4×10^{-25}
0.1	1.9×10^{-17}	3.3×10^{-19}	7.5×10^{-20}	6.0×10^{-22}	1.0×10^{-23}	2.4×10^{-24}
1	1.8×10^{-17}	3.2×10^{-19}	7.3×10^{-20}	5.8×10^{-21}	1.0×10^{-22}	2.3×10^{-23}
10	1.4×10^{-17}	2.4×10^{-19}	5.5×10^{-20}	4.4×10^{-20}	7.7×10^{-22}	1.7×10^{-22}
25	8.6×10^{-18}	1.5×10^{-19}	3.4×10^{-20}	6.7×10^{-20}	1.2×10^{-21}	2.7×10^{-22}
50	3.9×10^{-18}	6.8×10^{-20}	1.5×10^{-20}	6.1×10^{-20}	1.1×10^{-21}	2.4×10^{-22}
100	7.8×10^{-19}	1.4×10^{-20}	3.1×10^{-21}	2.5×10^{-20}	4.4×10^{-22}	9.8×10^{-23}
250	6.6×10^{-21}	1.2×10^{-22}	2.6×10^{-23}	5.2×10^{-22}	9.1×10^{-24}	2.1×10^{-24}
500	2.3×10^{-24}	4.0×10^{-26}	9.1×10^{-27}	3.6×10^{-25}	6.9×10^{-27}	1.4×10^{-27}
1000	2.8×10^{-31}	4.9×10^{-33}	1.1×10^{-33}	8.8×10^{-33}	1.5×10^{-33}	3.5×10^{-34}

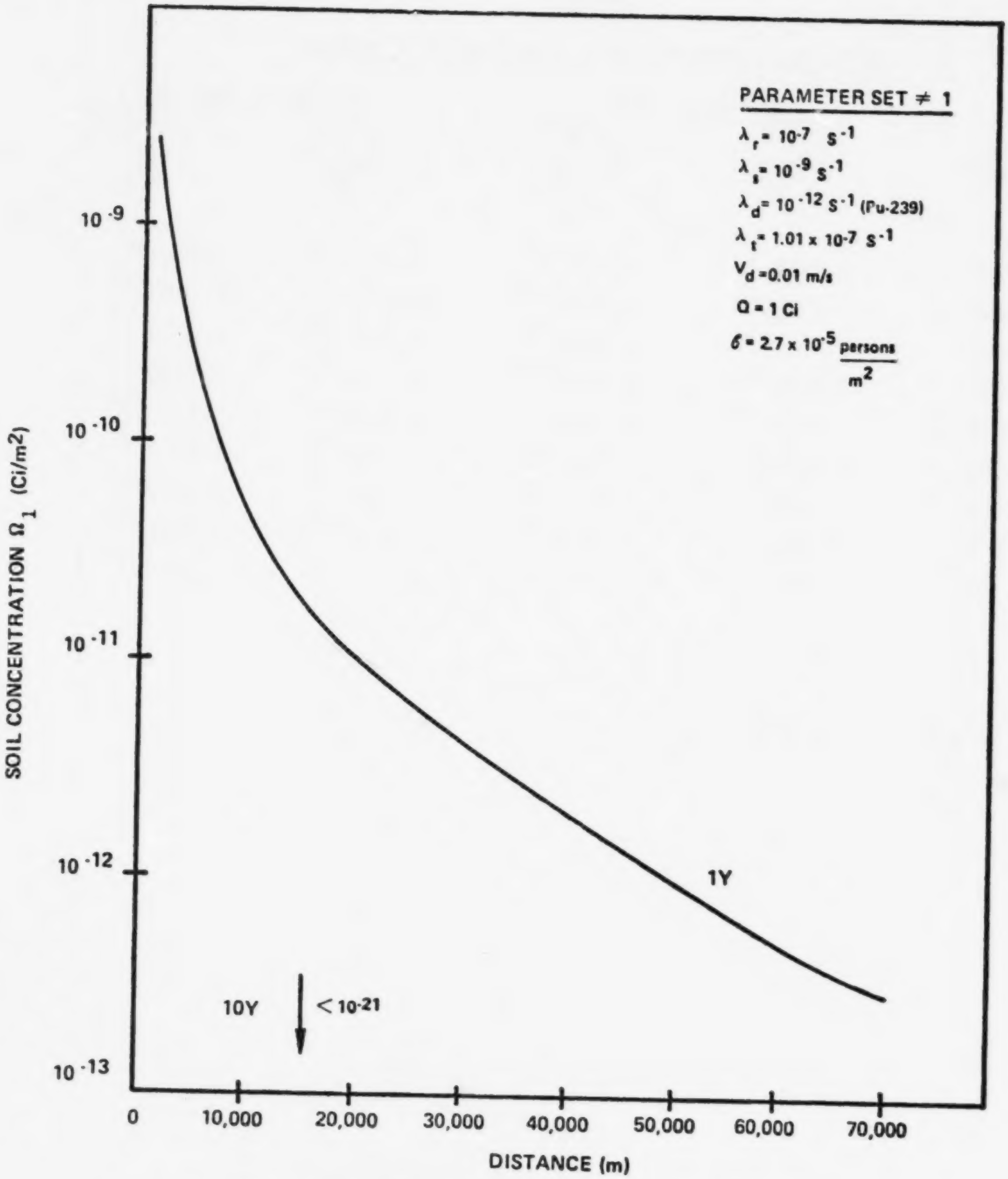
*corrected for deposition from plume

Table 9

Soil Concentration As A function Of Distance

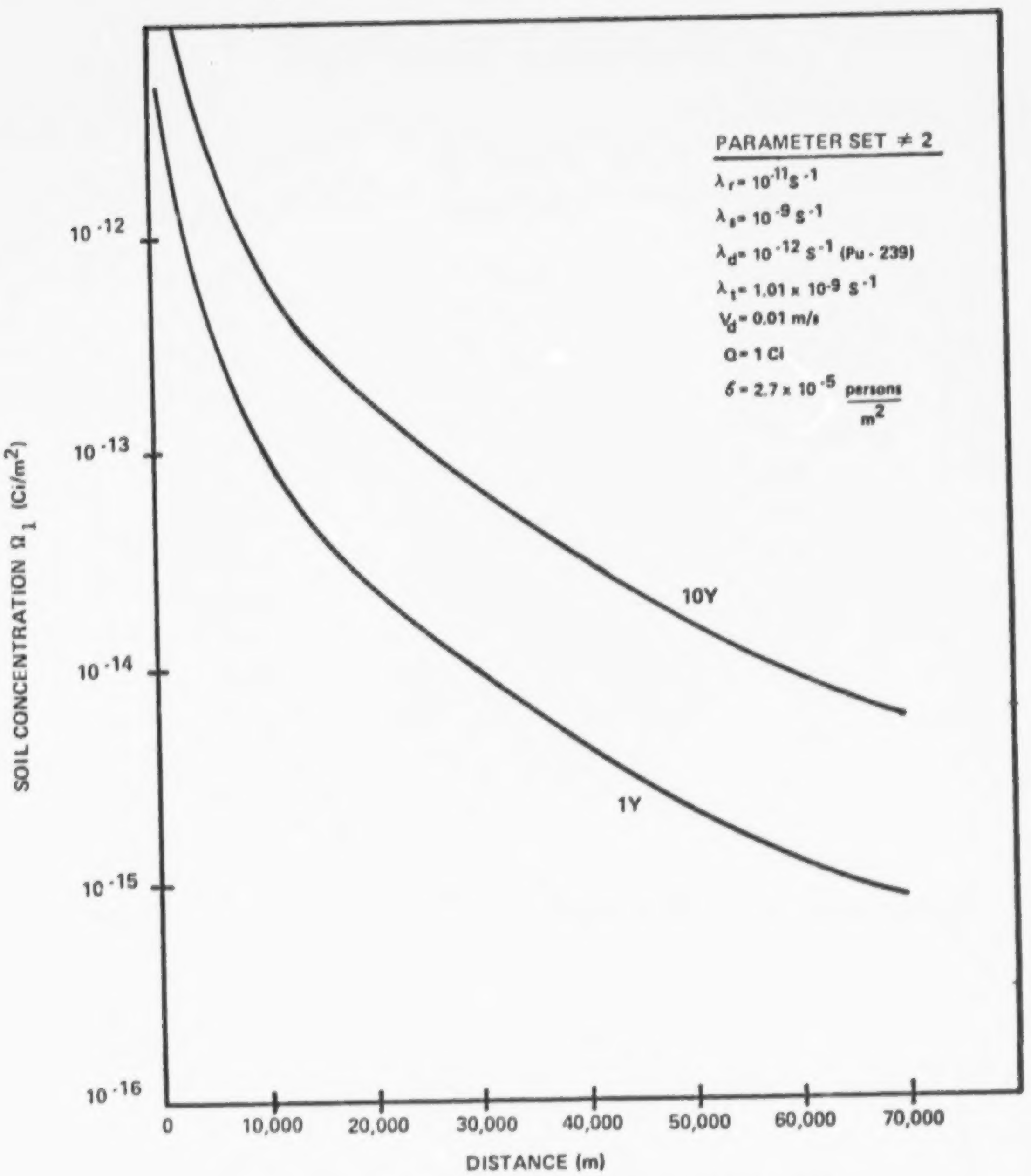
r	Parameter Set #1		Parameter Set #2	
	Ω_1(1y)	Ω_1(10y)	Ω_1(1y)	Ω_1(10y)
(m)	$(\frac{Ci}{m^2})$	$(\frac{Ci}{m^2})$	$(\frac{Ci}{m^2})$	$(\frac{Ci}{m^2})$
1,000	$2.5\text{x}10^{-9}$	$8.8\text{x}10^{-21}$	$5.8\text{x}10^{-12}$	$4.4\text{x}10^{-11}$
10,000	$4.4\text{x}10^{-11}$	$1.5\text{x}10^{-22}$	$1.0\text{x}10^{-13}$	$7.7\text{x}10^{-13}$
20,000	$9.8\text{x}10^{-12}$	$3.5\text{x}10^{-23}$	$2.3\text{x}10^{-14}$	$1.7\text{x}10^{-13}$
30,000	$3.7\text{x}10^{-12}$	$1.3\text{x}10^{-23}$	$8.7\text{x}10^{-15}$	$6.5\text{x}10^{-14}$
40,000	$1.7\text{x}10^{-12}$	$6.2\text{x}10^{-24}$	$4.1\text{x}10^{-15}$	$3.1\text{x}10^{-14}$
50,000	$9.4\text{x}10^{-13}$	$3.3\text{x}10^{-24}$	$2.2\text{x}10^{-15}$	$1.6\text{x}10^{-14}$
60,000	$5.4\text{x}10^{-13}$	$1.9\text{x}10^{-24}$	$1.3\text{x}10^{-15}$	$9.6\text{x}10^{-15}$
70,000	$3.4\text{x}10^{-13}$	$1.2\text{x}10^{-24}$	$7.9\text{x}10^{-16}$	$5.9\text{x}10^{-15}$
80,000	$2.2\text{x}10^{-13}$	$7.7\text{x}10^{-25}$	$5.1\text{x}10^{-16}$	$3.8\text{x}10^{-15}$

$$\Omega_1(r,t) = V_d \lambda_r Q \left(\frac{X}{\dot{Q}}\right)_{r_n} \left(\frac{r}{r_n}\right)^{-\ell} \exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right] t \ \exp[-\lambda_t t]$$



SOIL CONCENTRATION VS. DISTANCE – PARAMETER SET ≠ 1

FIGURE 7



SOIL CONCENTRATION VS. DISTANCE – PARAMETER SET ≠ 2
FIGURE 8

Table 10

Soil Concentration As A Function Of Time At Various Distances-Parameter Set #1

t	$\Omega_1(\frac{Ci}{m^2})$		
(y)	r=1,000m	r=10,000m	r=20,000m
0.01	5.8×10^{-10}	1.0×10^{-11}	2.3×10^{-12}
0.1	4.4×10^{-9}	7.7×10^{-11}	1.7×10^{-11}
0.2	6.3×10^{-9}	1.1×10^{-10}	2.5×10^{-11}
0.3	6.9×10^{-9}	1.2×10^{-10}	2.7×10^{-11}
0.4	6.7×10^{-9}	1.17×10^{-10}	2.66×10^{-11}
0.6	5.3×10^{-9}	9.3×10^{-11}	2.1×10^{-11}
0.8	3.7×10^{-9}	6.6×10^{-11}	1.5×10^{-11}
1	2.5×10^{-9}	4.4×10^{-11}	9.8×10^{-12}
10	8.8×10^{-21}	1.5×10^{-22}	3.5×10^{-23}
20	$<10^{-35}$	$<10^{-35}$	$<10^{-36}$
50	"	"	"
100	"	"	"
500	"	"	"
1000	"	"	"

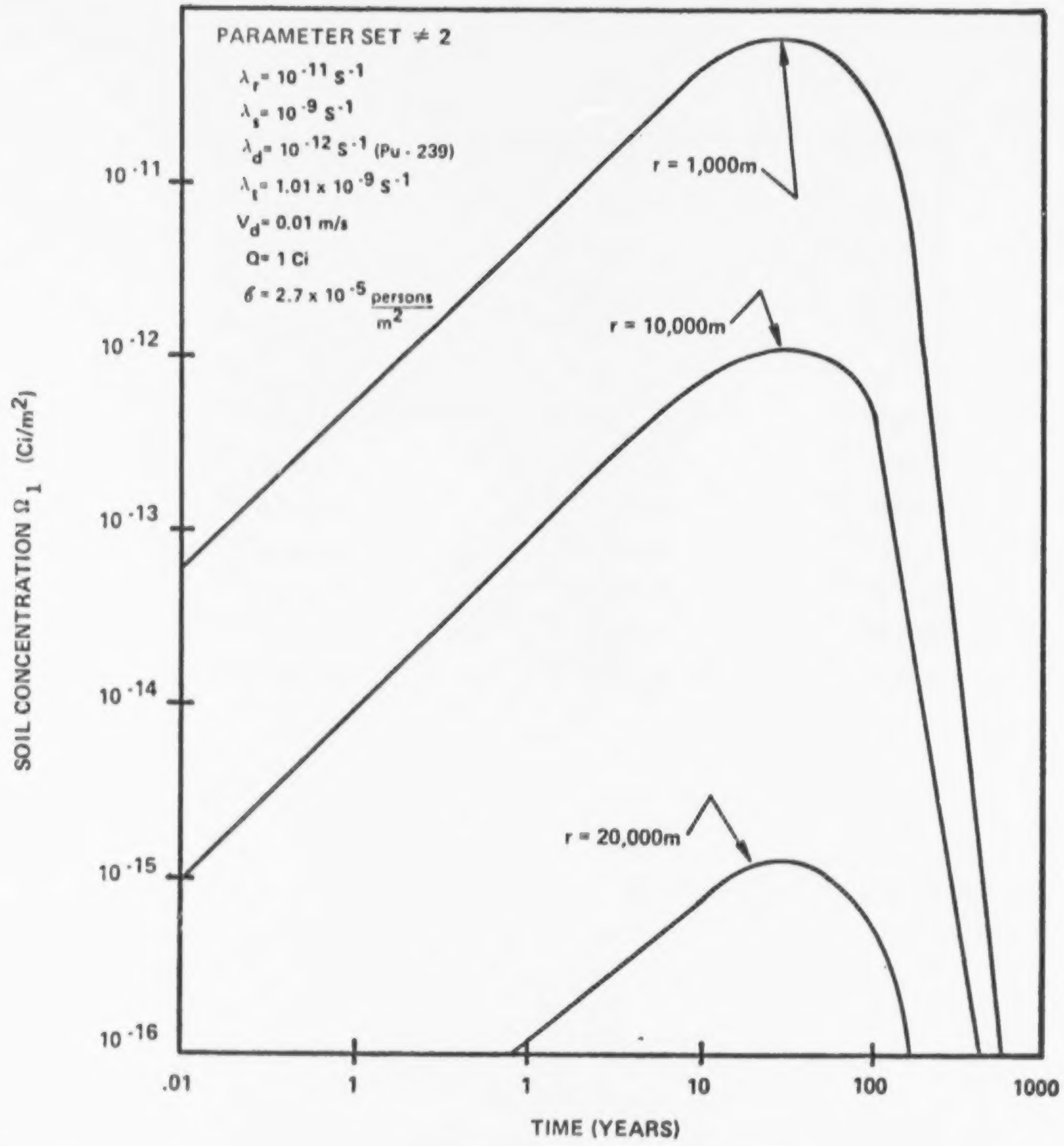
Ω_1(max) occurs at $t = \frac{1}{\lambda_t} = .314y$

Table 11

Soil Concentration As A Function Of Time At Various Distances-Parameter Set #2

t (y)	$\Omega_1(\frac{Ci}{m^2})$		
	r=1,000m	r=10,000m	r=20,000m
0.01	6.0×10^{-14}	1.1×10^{-15}	1.2×10^{-18}
0.1	6.0×10^{-13}	1.0×10^{-14}	1.2×10^{-17}
1	5.8×10^{-12}	1.0×10^{-13}	1.2×10^{-16}
10	4.4×10^{-11}	7.7×10^{-13}	8.9×10^{-16}
20	6.3×10^{-11}	1.1×10^{-12}	1.3×10^{-15}
35	6.9×10^{-11}	1.2×10^{-12}	1.4×10^{-15}
40	6.7×10^{-11}	1.17×10^{-12}	1.36×10^{-15}
50	6.1×10^{-11}	1.1×10^{-12}	1.2×10^{-15}
100	2.5×10^{-11}	4.4×10^{-13}	5.0×10^{-16}
250	5.2×10^{-13}	9.2×10^{-15}	1.1×10^{-17}
500	3.6×10^{-16}	6.4×10^{-18}	7.4×10^{-21}
1000	8.8×10^{-23}	1.5×10^{-24}	1.8×10^{-27}

Ω_1(max) occurs at $t = \frac{1}{\lambda_t} = 31.43y$



SOIL CONCENTRATION VS. TIME
FIGURE 9

APPENDIX I

Derivation Of $X/\dot{Q}$ Vs. Distance Relationship For A Ground Level Release

The former Atomic Energy Commission in its draft environmental statement for Appendix I, 10 CFR 50, (8) presented data on average annual atmospheric dilution factors, $\frac{X}{\dot{Q}}$, versus distance, r, at a ground level release height for 17 nuclear power reactor sites situated on rivers, lakes, and seacoasts. The data, plotted on a log-log graph, showed a linear relationship between the atmospheric dilution factor and distance. The general equation describing this line is:

$$\log_e y = m\log_e x + \log_e b \qquad (1)$$

$$\log_e y - \log_e b = m\log_e x$$

$$\log_e\left(\frac{y}{b}\right) = \log_e x^m$$

$$y = bx^m \qquad (2)$$

where m = slope of the line

b = value of y at $X = 1$

For a specific case: (see Fig. 1)

$$\frac{X}{\dot{Q}}(r) = br^m \qquad (3)$$

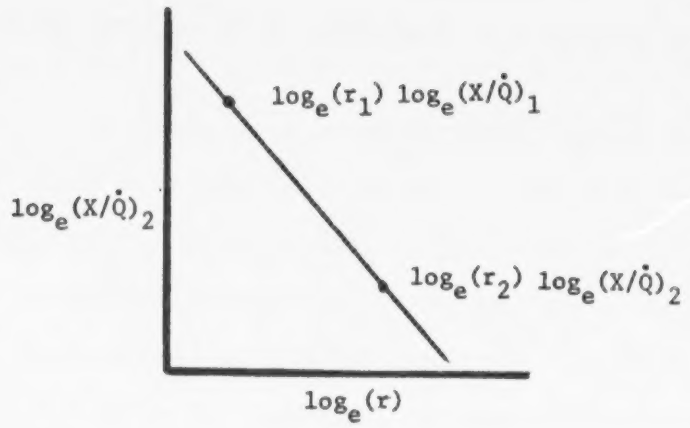
where

$\frac{X}{\dot{Q}}$ = average annual atmospheric dilution factor

r = distance

m = slope

Fig. 1


$\log_e(r_1)$ $\log_e(X/\dot{Q})_1$
$\log_e(X/\dot{Q})_2$
$\log_e(r_2)$ $\log_e(X/\dot{Q})_2$
$\log_e(r)$

The slope of the line in Fig. 1 is:

$$m = \frac{\Delta Y}{\Delta X} = \frac{\log_e(X/\dot{Q})_2 - \log_e(X/\dot{Q})_1}{\log_e(r_2) - \log_e(r_1)}$$

$$= \frac{\log_e\{(X/\dot{Q})_2/(X/\dot{Q})_1\}}{\log_e(r_2/r_1)}$$

$$m = \frac{\log_e\{(X/\dot{Q})_1/(X/\dot{Q})_2\}}{\log_e(r_2/r_1)} \qquad (4)$$

let $m = -\ell$

In equation (3), b is calculated for distances other than r=1 as follows. Select some arbitrary normalizing distance, r_n, and determine the corresponding $X/\dot{Q}$ value, $(X/\dot{Q})_{r_n}$, from Fig. 1. At $r=r_n$ equation (3) becomes:

$$(X/\dot{Q})_{r_n} = (b)_{r_n} r_n^{-\ell}$$

or $$(b)_{r_n} = \left(\frac{X}{\dot{Q}}\right)_{r_n} r_n^{\ell} \qquad (5)$$

Substituting equation (5) into equation (3):

$$\frac{X}{\dot{Q}}(r) = \left(\frac{X}{\dot{Q}}\right)_{r_n} r_n^{\ell} r^{-\ell}$$

$$\frac{X}{\dot{Q}}(r) = \left(\frac{X}{\dot{Q}}\right)_{r_n} \left(\frac{r}{r_n}\right)^{-\ell} \qquad (6)$$

Equation (6) describes the atmospheric dilution factor, $X/\dot{Q}$, without taking into account depletion of the plume. This is valid for noble gases, but not for particulate sources whose plume concentration is depleted by deposition of material as the plume moves downwind. A correction factor for depletion can be derived two ways, by depleting the plume as it travels downwind or by depleting source term. Both methods yield the same result. The ~~depletion correction~~ factor for this case will be derived by the method of depleting the source.

let $\frac{\dot{Q}^d}{\dot{Q}}$ = depletion correction factor

where $\dot{Q}$ = undepleted source term

$\dot{Q}^d$ = depleted source term

Correcting equation (6) for depletion and denoting the depleted $X/\dot{Q}$ as $X^d/\dot{Q}^d$:

$$\frac{X^d}{\dot{Q}^d}(r) = \left(\frac{X}{\dot{Q}}\right)_{r_n} \left(\frac{r}{r_n}\right)^{-\ell} \left(\frac{\dot{Q}^d}{\dot{Q}}\right) \qquad (7)$$

The depleted source term $\dot{Q}^d$ is derived as follows:

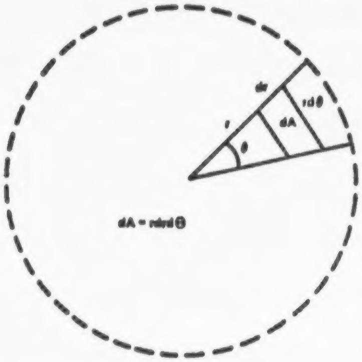
Assume the deposition rate, $\omega(r)$, at a distance r to be:

$$\omega(r) = XV_d$$

where X = undepleted air concentration

V_d = deposity velocity

Fig. 2



The depletion of a differential source term $d\dot{Q}$ in area, dA, using polar coordinates, is:

$$d\dot{Q} = -XV_d dA$$

$$= XV_d rdrd\Theta \qquad (8)$$

from equation (6)

$$X = \dot{Q}\left(\frac{X}{\dot{Q}}\right)_{r_n}\left(\frac{r}{r_n}\right)^{-\ell}$$

Substituting into equation (8) and separating variables:

$$\frac{d\dot{Q}}{\dot{Q}} = - \left(\frac{X}{\dot{Q}}\right)_{r_n} \left(\frac{r}{r_n}\right)^{-\ell} V_d r dr d\Theta$$

Integrating over 0 to r and 0 to 2π

$$\int_0^r \frac{d\dot{Q}}{\dot{Q}} = - \left(\frac{X}{\dot{Q}}\right)_{r_n} V_d \int_0^r \int_0^{2\pi} r\left(\frac{r}{r_n}\right)^{-\ell} dr d\Theta$$

$$\log_e(\dot{Q})\Big|_0^r = - \left(\frac{X}{\dot{Q}}\right)_{r_n} V_d 2\pi r_n^{-\ell} \int_0^r r^{1-\ell} dr$$

$$\log_e \dot{Q}(r) - \log_e \dot{Q}(0) = - 2\pi V_d r_n^{\ell} \left(\frac{X}{\dot{Q}}\right)_{r_n} \left[\frac{r^{2-\ell}}{2-\ell}\right]$$

let $\dot{Q}(0) = \dot{Q}$

$\dot{Q}(r) = \dot{Q}^d$

$$\log_e\left(\frac{\dot{Q}^d}{\dot{Q}}\right) = - \frac{2\pi V_d}{2-\ell} \left(\frac{X}{\dot{Q}}\right)_{r_n} \frac{r^{2-\ell}}{r_n^{-\ell}} \qquad (9)$$

$$\text{let } r_d = \left[\frac{(2-\ell) r_n^{-\ell}}{2\pi V_d \left(\frac{X}{\dot{Q}}\right)_{r_n}}\right]^{\frac{1}{2-\ell}}$$

Therefore:

$$\left(\frac{1}{r_d}\right)^{2-\ell} = \frac{2\pi V_d}{2-\ell} \left(\frac{X}{\dot{Q}}\right)_{r_n} \left(\frac{1}{r_n^{-\ell}}\right)$$

Substituting into equation (9)

$$\log_e\left(\frac{\dot{Q}^d}{\dot{Q}}\right) = -\left(\frac{1}{r_d}\right)^{2-\ell} r^{2-\ell}$$

$$\log_e\left(\frac{\dot{Q}^d}{\dot{Q}}\right) = -\left(\frac{r}{r_d}\right)^{2-\ell}$$

and

$$\frac{\dot{Q}^d}{\dot{Q}} = \exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right] \tag{10}$$

Substituting this into equation (8), the depleted $X/\dot{Q}$ equation becomes:

$$\frac{X^d}{\dot{Q}^d}(r) = \left(\frac{X}{\dot{Q}}\right)_{r_n} \left(\frac{r}{r_n}\right)^{-\ell} \exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right] \tag{11}$$

In equation (10), r_d represents a deposition distance, when $r=r_d$, $\frac{\dot{Q}^d}{\dot{Q}}$ has decreased by a factor of $\frac{1}{e}$ or 37%.

The AEC data presented in reference (8) can be used to obtain an expression for the undepleted $X/\dot{Q}$ as given in equation (6).

$$\frac{X}{\dot{Q}}(r) = \left(\frac{X}{\dot{Q}}\right)_{r_n} \left(\frac{r}{r_n}\right)^{-\ell}$$

let $r_n = 1$ km

$\frac{X}{\dot{Q}}$ at 1 km varies from 2 to $3\times10^{-6}\ \frac{s}{m^3}$

assume $\frac{X}{\dot{Q}}$ at 1 km = $2.5\times10^{-6}\ \frac{s}{m^3}$

$\frac{X}{\dot{Q}}$ at 160 km varies from 1.5 to $2.1\mathrm{x}10^{-9}\ \frac{s}{m^3}$

assume $\frac{X}{\dot{Q}}$ at 160 km = $1.8\mathrm{x}10^{-9}\ \frac{s}{m^3}$

The slope of the line is:

$$m = -\ell = -\frac{\log_e\{(X/\dot{Q})_{km}/(X/\dot{Q})_{160\ km}\}}{\log_e\left(\frac{160}{1}\right)}$$

$$-\ell = -\frac{\log_e\{2.5\mathrm{x}10^{-6}/1.8\mathrm{x}10^{-9}\}}{\log_e(160)}$$

$$\ell = 1.43$$

and $\left(\frac{X}{\dot{Q}}\right)_{1\ km} = 2.5\mathrm{x}10^{-6}\ \frac{s}{m^3}$

Therefore:

$$\frac{X}{\dot{Q}}(r) = 2.5\mathrm{x}10^{-6}\ r^{-1.43} \qquad (12)$$

This equation is the result of averaging the average annual atmospheric dilution factors for rivers, lakes, and seacoasts sites. It is valid only for those distances for which data was presented, i.e., .1 km<r<160 km. However data beyond 80 km is not very accurate.

Appendix II

Solutions To Selected Integrals And Differential Equations

A. Integration of Equation (13)

$$PE_0(r,t) = 2\pi\rho\lambda_r Q\left(\frac{X}{\dot{Q}}\right)_{r_n} \int_{t_1}^{t_2} \int_{r_1}^{r_2} r\left(\frac{r}{r_n}\right)^{-\ell} \exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right] \exp[-\lambda_t t]\,dr\,dt$$

Integrating over r first:

$$\int_{r_1}^{r_2} r\left(\frac{r}{r_n}\right)^{-\ell} \exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right] dr = \int_{r_1}^{r_2} \frac{r^{1-\ell}}{r_n^{-\ell}} \exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right] dr$$

By substitution:

$$\text{let } u = r^{2-\ell}$$

$$du = (2-\ell) r^{1-\ell} dr$$

$$u_0 = r_d^{2-\ell}$$

The new limits of integration are:

$$u_1 = r_1^{2-\ell}$$

$$u_2 = r_2^{2-\ell}$$

Therefore:

$$\int_{r_1^{2-\ell}}^{r_2^{2-\ell}} \left(\frac{r^{1-\ell}}{r_n^{-\ell}}\right)\left(\frac{du}{(2-\ell)r^{1-\ell}}\right)\exp\left(-\frac{u}{u_0}\right) = \frac{1}{r_n^{-\ell}(2-\ell)} \int_{r_1^{2-\ell}}^{r_2^{2-\ell}} \exp\left(-\frac{u}{u_0}\right) du$$

$$= \frac{u_0}{r_n^{-\ell}(2-\ell)} \left[-\exp\left(-\frac{u}{u_0}\right)\right]_{r_1^{2-\ell}}^{r_2^{2-\ell}}$$

$$= \frac{r_d^{\,2-\ell}}{r_n^{-\ell}(2-\ell)}\left[\exp\left[-\left(\frac{r_1}{r_d}\right)^{2-\ell}\right] - \exp\left[-\left(\frac{r_2}{r_d}\right)^{2-\ell}\right]\right]$$

$PE_0(r,t)$ now becomes:

$$PE_0(r,t) = 2\pi\rho Q\lambda_r\left(\frac{X}{\dot{Q}}\right)_{r_n}\frac{r_d^{\,2-\ell}}{r_n^{-\ell}(2-\ell)}\left[\exp\left[-\left(\frac{r_1}{r_d}\right)^{2-\ell}\right]-\exp\left[-\left(\frac{r_2}{r_d}\right)^{2-\ell}\right]\right]\int_{t_1}^{t_2}\exp(-\lambda_t t)dt$$

Integrating over t:

$$\int_{t_1}^{t_2}\exp(-\lambda_t t)dt = \frac{1}{\lambda_t}[\exp(-\lambda_t t_1)-\exp(-\lambda_t t_2)]$$

Also $$r_d^{2-\ell} = \left[\left(\frac{(2-\ell)(r_n^{-\ell})}{2\pi V_d\left(\frac{X}{\dot{Q}}\right)_{r_n}}\right)^{\frac{1}{2-\ell}}\right]^{2-\ell} = \frac{(2-\ell)r_n^{-\ell}}{2\pi V_d\left(\frac{X}{\dot{Q}}\right)_{r_n}}$$

Substituting in the time integration and the result of $r_d^{2-\ell}$ and simplfying gives:

$$PE_0(r,t) = \frac{\rho Q}{V_d}\left(\frac{\lambda_r}{\lambda_t}\right)\left[\exp\left[-\left(\frac{r_1}{r_d}\right)^{2-\ell}\right]-\exp\left[-\left(\frac{r_2}{r_d}\right)^{2-\ell}\right]\right][\exp(-\lambda_t t_1)-\exp(-\lambda_t t_2)]$$

B. Solution to Equation (17)

$$\dot{\Omega}_1(r,t) = -\lambda_t\Omega_1(r,t) + \omega_1(r,t)$$

or $$\dot{\Omega}_1(r,t) + \lambda_t\Omega_1(r,t) = \omega_1(r,t)$$

This linear first order differential equation is solved, with respect to time, by use of an integrating factor u(t), where:

$$u(t) = \exp\left[\int^t p(x)dx\right]$$

and $y = \frac{1}{u(t)}[\int^t u(s)p(s)ds + c]$ c = constant

where y is the solution of $\dot{y} + p(t)y = g(t)$

$$\omega_1(r,t) = V_d X_0^d(r,t)$$

substituting equation (8) for X_0^d

$$\omega_1(r,t) = V_d \lambda_r Q(\frac{X}{\dot{Q}})_{r_n} (\frac{r}{r_n})^{-\ell} \exp[-(\frac{r}{r_d})^{2-\ell}] \exp[-\lambda_t t]$$

Therefore:

$$\dot{\Omega}_1(r,t) + \lambda_t \Omega_1(r,t) = V_d \lambda_r Q(\frac{X}{\dot{Q}})_{r_n} (\frac{r}{r_n})^{-\ell} \exp[-(\frac{r}{r_d})^{2-\ell}] \exp[-\lambda_t t]$$

let $A = V_d \lambda_r Q(\frac{X}{\dot{Q}})_{r_n} (\frac{r}{r_n})^{-\ell} \exp[-(\frac{r}{r_d})^{2-\ell}]$

$$\dot{\Omega}_1(r,t) + \lambda_t \Omega_1(r,t) = A \exp[-\lambda_t t]$$

The integrating factor u(t) is:

$$u(t) = \exp[\int^t p(x)dx] = \exp[\int^t \lambda_t dx]$$

$$u(t) = \exp[\lambda_t t]$$

and

$$\Omega_1(r,t) = \frac{1}{u(t)} [\int^t u(s)g(s)ds + c]$$

$$\Omega_1(r,t) = \exp[-\lambda_t t][\int^t \exp[\lambda_t t](A \exp[-\lambda_t t]ds + c]$$

$$\Omega_1(r,t) = \exp[-\lambda_t t][\int^t Ads + c]$$

$$\Omega_1(r,t) = At\exp[-\lambda_t t] + c\exp[-\lambda_t t]$$

$$\Omega_1(0) = 0 \text{ therefore } c = 0$$

$$\Omega_1(r,t) = At\exp[-\lambda_t t] \qquad \text{substituting for A}$$

$$\Omega_1(r,t) = V_d \lambda_r Q(\frac{X}{\dot{Q}})_{r_n} (\frac{r}{r_n})^{-\ell} \exp[-(\frac{r}{r_d})^{2-\ell}] t\exp(-\lambda_t t)$$

C. Integration of Equation (29)

$$PE_1(r,t) = 2\pi\rho\lambda_r^2 Q(\frac{X}{\dot{Q}})_{r_n} \int_{t_1}^{t_2} \int_{r_1}^{r_2} r(\frac{r}{r_n})^{-\ell} \exp[-(\frac{r}{r_d})^{2-\ell}] t\exp[-\lambda_t t] dr dt$$

Integrating over r first and using result of part A in this Appendix

$$\int_{r_1}^{r_2} r(\frac{r}{r_n})^{-\ell} \exp[-(\frac{r}{r_d})^{2-\ell}] dr = \frac{r_d^{2-\ell}}{r_n^{-\ell}(2-\ell)} [\exp[-(\frac{r_1}{r_d})^{2-\ell}] - \exp[-(\frac{r_2}{r_d})^{2-\ell}]]$$

$PE_1(r,t)$ now becomes:

$$PE_1(r,t) = 2\pi\rho\lambda_r^2 Q(\frac{X}{\dot{Q}})_{r_n} \frac{r_d^{2-\ell}}{(2-\ell)r_n^{-\ell}} [\exp[-(\frac{r_1}{r_d})^{2-\ell}] - \exp[-(\frac{r_2}{r_d})^{2-\ell}]] \int_{t_1}^{t_2} t\exp(-\lambda_t t) dt$$

Integrating time integral by parts:

$$\int u dv = uv - \int v du$$

$$\int_{t_1}^{t_2} t\exp(-\lambda_t t)dt \quad \text{let} \quad u = t \quad du = \exp[-\lambda_t t]dt$$

$$du = dt \quad v = \frac{1}{\lambda_t}\exp(-\lambda_t t)$$

$$\int_{t_1}^{t_2} t\exp(-\lambda_t t)dt = -\frac{t}{\lambda_t}\exp(-\lambda_t t)\Big|_{t_1}^{t_2} + \frac{1}{\lambda_t}\int_{t_1}^{t_2}\exp[-\lambda_t t]dt$$

$$= \frac{t_1}{\lambda_t}\exp[-\lambda_t t_1] - \frac{t_2}{\lambda_t}\exp[-\lambda_t t_2] + \frac{1}{\lambda_t^2}\exp[-\lambda_1 t_1] - \frac{1}{\lambda_t^2}\exp[-\lambda_t t_2]$$

Combining terms:

$$\int_{t_1}^{t_2} t\exp(-\lambda_t t)dt = \left(\frac{t_1}{\lambda_t} + \frac{1}{\lambda_t^2}\right)\exp(-\lambda_t t_1) - \left(\frac{t_2}{\lambda_t} + \frac{1}{\lambda_t^2}\right)\exp(-\lambda_t t_2)$$

$$= \left(\frac{\lambda_t t_1 + 1}{\lambda_t^2}\right)\exp(-\lambda_t t_1) - \left(\frac{\lambda_t t_2 + 1}{\lambda_t^2}\right)\exp(-\lambda_t t_2)$$

Substituting in the time integration and the result of $r_d^{2-\ell}$ and simplifying gives:

$$PE_1(r,t) = \frac{\rho Q\lambda_r^2}{V_d}[\exp[-(\frac{r_1}{r_d})^{2-\ell}] - \exp[-(\frac{r_2}{r_d})^{2-\ell}]$$

$$[(\frac{\lambda_t t_1 + 1}{\lambda_t^2})\exp(-\lambda_t t_1) - (\frac{\lambda_t t_2 + 1}{\lambda_t^2})\exp(-\lambda_t t_2)]$$

Appendix III

Summary of Equations

(1) $$X_0^d(r,t) = \lambda_r Q\left(\frac{X}{\dot{Q}}\right)_{r_n}\left(\frac{r}{r_n}\right)^{-\ell} \exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right] \exp[-\lambda_t t]$$

$X_0^d(r,t)$ = depleted air concentration at distance r and time t resulting from dispersion of resuspended material from source $(\frac{Ci}{m^3})$

(2) $$X_1^d(r,t) = \lambda_r^2\, Q\left(\frac{X}{\dot{Q}}\right)_{r_n}\left(\frac{r}{r_n}\right)^{-\ell} \exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right] t\exp[-\lambda_t t]$$

$X_1^d(r,t)$ = depleted air concentration at distance r and time t resulting from resuspension of material that deposited out from X_0^d $(\frac{Ci}{m^3})$

(3) $$PE_0(r,t) = \frac{\rho Q}{V_d}\left(\frac{\lambda_r}{\lambda_t}\right)\left[\exp\left[-\left(\frac{r_1}{r_d}\right)^{2-\ell}\right] - \exp\left[-\left(\frac{r_2}{r_d}\right)^{2-\ell}\right]\right]\left[\exp(-\lambda_t t_1) - \exp(-\lambda_t t_2)\right]$$

$PE_0(r,t)$ = population inhalation exposure due to air concentration X_0^d (person - $\frac{Ci}{m^3}$ - s)

(4) $$PE_0(\infty,\infty) = \frac{\rho Q}{V_d}\left(\frac{\lambda_r}{\lambda_t}\right)$$

$PE_0(\infty,\infty)$ = the infinite population inhalation exposure,
$r = 0 \to \infty$, $t = 0 \to \infty$ (person - $\frac{Ci}{m^3}$ - s)

(5) $$PE_1(r,t) = \frac{\rho Q \lambda_r^2}{V_d}\left[\exp\left[-\left(\frac{r_1}{r_d}\right)^{2-\ell}\right] - \exp\left[-\left(\frac{r_2}{r_d}\right)^{2-\ell}\right]\right]$$

$$\left[\left(\frac{\lambda_t t_1 + 1}{\lambda_t^2}\right)\exp(-\lambda_t t_1) - \left(\frac{\lambda_t t_2 + 1}{\lambda_t^2}\right)\exp(-\lambda_t t_2)\right]$$

$PE_1(r,t)$ = population inhalation exposure due to air
concentration X_1^d (person - $\frac{Ci}{m^3}$ - s)

(6) $$PE_1(\infty,\infty) = \frac{\rho Q}{V_d}\left(\frac{\lambda_r}{\lambda_t}\right)^2$$

$PE_1(\infty,\infty)$ = the infinite population inhalation exposure
$r = 0 \to \infty$, $t = 0 \to \infty$ (person - $\frac{Ci}{m^3}$ - s)

(7) $$PE_T(\infty,\infty) = \frac{\rho Q}{V_d}\left(\frac{\lambda_r}{\lambda_d+\lambda_s}\right)$$

$PE_t(\infty,\infty)$ = the infinite total population inhalation exposure due to all resuspensions, $r = 0 \to \infty$, $t = 0 \to \infty$ (person - $\frac{Ci}{m^3}$ - s)

(8) $$\Omega_1(r,t) = V_d \lambda_r Q\left(\frac{X}{\dot{Q}}\right)_{r_n}\left(\frac{r}{r_n}\right)^{-\ell} \exp\left[-\left(\frac{r}{r_d}\right)^{2-\ell}\right] t \exp[-\lambda_t t]$$

$\Omega_1(r,t)$ = soil surface concentration at distance r and time t $\left(\frac{Ci}{m^2}\right)$

(9) $$r_2 = r_d[-\ell n(1-X)]^{\frac{1}{2-\ell}}$$

r_2 = the distance from the point source within which the fraction X of the pop. exposure is delivered (m)

(10) $$X = 1-\exp[-(\frac{r_2}{r_d})^{2-\ell}]$$

X = fraction of population inhalation exposure delivered within distance r_2

(11) $$r_d = [\frac{(2-\ell)\, r_n^{-\ell}}{2\pi V_d (X/\dot{Q})_{r_n}}]^{\frac{1}{2-\ell}}$$

r_d = a depletion distance, when $r = r_d$ the atmospheric dilution factor $\frac{Q^d}{\dot{Q}}(r)$ has decreased by a factor of 1/e or 37% (m)

Q = activity of point source (Ci)

r = distance from receptor to point source (m)

$(\frac{X}{\dot{Q}})_{r_n}$ = value of $\frac{X}{\dot{Q}}(r)$ at $r = r_n$ $(\frac{sec}{m^3})$

r_n = normalizing distance (m)

λ_r = resuspension rate (s^{-1})

λ_s = transfer rate of contaminated material from soil surface to soil sink (s^{-1})

λ_d = radiological decay constant (s^{-1})

$\lambda_t = \lambda_r + \lambda_s + \lambda_d$ (s^{-1})

ρ = population density (persons/m^2)

V_d = deposition velocity (m/sec)

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THE PHYSIOLOGICAL BASIS
OF TRANSURANIC ELEMENT
DOSE ESTIMATES

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THE PHYSIOLOGICAL BASIS OF TRANSURANIC ELEMENT DOSE ESTIMATES

Despite extensive literature reviews, scientific information on the biological properties of transuranium elements in humans is sparse and the medical prognosis following inhalation or ingestion of these substances is uncertain. Because of the care taken in handling plutonium by those groups who work with it and the length of the latent period before cancer develops, little information is available or likely to become available in the near future on the biological effects of plutonium in man. The basis for establishing metabolic parameters in humans is even less promising, since experiments using hospitalized patients are no longer performed.

HUMAN STUDIES

In 1945-46, 18 hospitalized persons were injected with tracer amounts of plutonium (1). Fifteen of the patients were 45 years of age or older. However, three had bone disorders, either fractures or cancer; three had liver disorders, two had kidney disorders, and the rest had other conditions which may have affected their metabolism of plutonium. Only about six of the persons studied provided reasonable data in that they were "normal" for most of the metabolic parameters examined (1).

To some extent supportive studies on metabolism of plutonium have been done using other groups of humans, but, because members of these other groups were accidentally exposed and the initial dose is not known with any degree of accuracy, about the only information available from these groups is confirmation of trends of metabolism and effective half-lives. The accidentally exposed groups are being followed both for the metabolic information and for information on possible health effects.

The groups being followed to develop information on the long-term or latent effects of plutonium exposure in man include:

1. Groups presently studied at Los Alamos Scientific Laboratory (2)

   a. The UPPU* group, 25 men exposed during 1944-45 with relatively large doses, (exposed primarily by inhalation; about 75% have one or more maximum permissible body burdens (3)).

   b. A group of 42 men exposed during the Manhattan Project (1943-1946), (exposed primarily by inhalation; have less than one maximum permissible body burden (3).

   c. A group of 190 early and current plutonium workers, (about 75% exposed by inhalation, 25% through wounds; about 20% have one or more maximum permissible body burdens (2)).

2. The United States Transuranium Registry

The U.S. Transuranium Registry was established in 1968 to protect the interests of workers, employees and the public by serving as a national focal point for acquisition and provision of information about the effects of transuranic elements on man. In October 1975, the Registry had identified 9063 transuranium workers and obtained autopsies on 53 of them. Of the workers identified, about 60% have burdens less than 5% of maximum permissible body burdens (4). Thus, the data base for the effects of plutonium in man is less than 10,000 exposed persons, most of whom were only nominally exposed and most of whom are alive today.

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*UPPU is an acronym of: U = Pronoun; P = Urinate; Pu = Plutonium.

As Thompson (5) has pointed out, the histological changes at the cellular level that have been observed in man following plutonium exposure cannot be qualitatively related to health consequences. Therefore, we know nothing directly about effects of plutonium in humans. Thompson (5) suggested since we have no useful data on human plutonium effects, it is useful 1) to extrapolate from plutonium effects in animals to plutonium effects in man or 2) to extrapolate from nonplutonium radiation effects in man to plutonium effects in man. He felt that not only was there "ballpark" agreement between human and animal data, where direct human data could be compared to experimental animal data, but also that plutonium data from animals could reasonably be extrapolated to man.

In 1976, Thompson (6) reiterated his plea for use of animal data suggesting that it is unlikely that anything useful about dose effect relationships will be learned from humans exposed to plutonium. He stated, however, that it is worthwhile to continue to study human exposure cases to obtain information on metabolism and to insure that there are no "surprises" in the biological effects of plutonium in man (6).

ANIMAL STUDIES

The literature on plutonium and the transuranic elements is voluminous. Starting with some of the earliest reviews of the subject (7-9) and speciality symposia (10) and continuing until the present (11-19) the emphasis has been primarily on animal experiments as the source of data on metabolism and hazards of plutonium. The data, based on animal studies on the distribution and metabolism of plutonium, have been summarized in ICRP-19 (20) and used by some to calculate doses in rads. The ICRP recommends that the numerical values derived for plutonium deposition in the various organs be applied to all transuranics. At best this is only a first approximation since the metabolic characteristics of the transuranics do differ.

A comparison of the organ distribution of plutonium in several species of mammals indicates that the soluble plutonium distribution estimates of 45% in the skeleton, 45% in the liver, and 10% in soft tissues proposed some years ago in ICRP-19 (20), are more likely to be 43% in the skeleton, 34% in the liver, 5% in bone marrow and 19% in soft tissues and excreta (21,22). In addition, on the basis of animal data, the distribution of soluble americium and curium would be estimated as: skeleton - 35% and 25%, liver - 57% and 60%, bone marrow - 4% and 3%, and soft tissue and excreta - 4% and 12%, respectively (21).

The Agency employs both animal and human data bases in deriving health effects estimates for plutonium, using extrapolation 1) from animals to man and 2) of nonplutonium radiation effects to plutonium effects in man. Animal data are used primarily to estimate distribution factors and retention parameters, and to relate these to the sparse human data. Following the NAS-BEIR Committee recommendations, human data are used primarily for dose-response conversion estimates (23). EPA has noted that as the Nuclear Regulatory Commission pointed out (24) there are great difficulties in extrapolating dose-response data between strains of the same species and even greater difficulties extrapolating between species. Therefore, the Agency agrees that major reliance on human dose-response data appears to be the most prudent course. Since there are so little data on transuranic element metabolism in man, extrapolation must be made from animal data. But, the extrapolation must be on the basis of what is observed in several diverse species rather than in a single species.

MODELS FOR TRANSURANIC DOSIMETRY

Inhalation

The model currently used by the EPA Office of Radiation Programs for estimating deposition and retention of inhaled transuranics in man is the ICRP Task Group on Lung Dynamics [TGLD] model (25) as modified by ICRP Publication #19 (20).

In this model, three chemical classes of compounds are considered based on the rate of elimination from the lung; Class Y compounds in years, Class W in weeks and Class D in days. Class Y compounds include carbides, oxides, hydroxides and lanthanide fluorides.* Class W compounds include nitrates, carbonates and lanthanide halides and phosphates. Class D compounds include all highly soluble materials. Information is not available on all plutonium compounds, but the ready hydrolysis of uncomplexed actinides suggests that no actinide compound would be in Class D.

The parameters used by the TGLD for estimating fractional deposition within regions of the lung for particles of differing "activity median aerodynamic diameter" (AMAD)** are based on a 30 year old adult standard male, breathing through the nose at a rate of 15 respirations per minute and with a tidal volume 0f 750 cm^3, 1450 cm^3 or 2150 cm^3. Retention expected in the lungs would be:

Lung -- For inhaled Class Y compounds, 13.8% of the inhaled material is retained in the pulmonary region of the lung, with a 500 day half-life. For Class W compounds, the 13.8% of the inhaled material is retained with a 50-day half-life. In the tracheobronchial region of the lung clearance half-times are 0.2 days or less; in the nasopharyngeal region they are 0.4 days or less (20).

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*Rapid translocation of curium oxide from the dog lung suggests the possibility of exceptions to this Class Y retention for some actinide compounds.

**The Aerodynamic Diameter (AD, Aerodynamic Equivalent Diamter) is defined as the diameter of a unit density sphere, which has the same settling speed under gravity as the particle being described. The Activity Median Diameter (AMD) is the diameter of the particles at the median of the log normal distribution of the radioactivity in an aerosol. The AMAD, then is the diameter of a unit density sphere having the same settling properties under gravity as the AMD particles.

Lymph Nodes -- About 3.4% of the Class Y material deposited in the lung is translocated to the thoracic lymph nodes and retained with a half-life of 1000 days. In Class W, 1.15% of the deposited material is retained with a half-life of 50 days (20).

The ORP inhalation code PAID (26) is based on the Task Group on Lung Dynamics model with appropriate adjustments for continuous intake of airborne radioactive materials. The PAID code employs ICRP-23 estimates of eight hours at rest (tidal volume 500 cm^3) and eight hours each of "light" and nonoccupational activity (tidal volume 1250 cm^3) as the basis of calculations (27).

The burden, $q_{k\ell}$, due to chronic inhalation, for any of the ℓ ICRP lung subcompartments is given by

$$\sum_{\ell} q_{k\ell}(t) = \frac{I\, f_{\ell}\, D_{k}\, (1 - e^{-\lambda_{\ell} t})}{\lambda_{\ell}}$$

where I = intake rate (pCi/y)

f_{ℓ} = fraction of material clearing by the ℓth pathway.

D_{k} = deposition fraction for kth compartment

λ_{ℓ} = effective half-life for the ℓth subcompartment.

The total lung burden is found by summing this equation over all subcompartments. For organs receiving plutonium subsequent to its transport from the lung, the equations are somewhat more complicated since each pathway must be traced.

The cumulative organ burden is then defined as

$$Q(t) = \int_0^T q(t)\,dt$$

In principle, this cumulative organ burden can be used with appropriate dose-conversion factors to calculate the annual dose rate to the organ in rad per year. The annual organ dose rate in turn can be used to estimate health effects, e.g., malignancy in either terms of individual risk or population risk. In actual practice the uncertainty in such risk estimates makes these calculations, at best, only estimates of expected response.

Ingestion of Inhaled Transuranics

Some of the inhaled transuranics will be cleared from the lung via the mucus escalator, swallowed, transported to the GI tract just as if they had been ingested. The calculation of transport from the lung to the GI tract indicates that about 92% of inhaled Class Y material will enter the GI tract, and about 81% of Class W material. Table I lists the relative disposition of inhaled material by region for a nominal 1.0 μ AMAD aerosol, assumed in reference 25.

Distribution of Transuranics in the Body

The material which has been transported into the blood stream will distribute in the organs of the body. ICRP estimates the following distributions:

Skeleton -- 45% of the transuranics in the circulatory system is expected to deposit in the skeleton and be retained with a half-life of about 100 years (20). In the case of very stable, biologically inactive complexes of plutonium (e.g., plutonium-DTPA) as little as 10% may deposit in the skeleton.

Liver -- 45% of the transuranics in the circulatory system may be deposited in the liver and retained with a half-life of 40 years (20). As in the case of the skeleton only 10% of stable, biologically inactive complexes may deposit in the liver.

Soft Tissues -- About 7% of the plutonium in the circulatory system may be deposited in soft tissue in the spleen, ovaries, uterus, testis, and adrenal glands. [From data in references 1 and 28, the 10% in soft tissue and excreta estimated by ICRP (20) may be divided into 7% and 3%, respectively.] Plutonium deposits are retained for extended periods with half-lives of 1500 days or longer. However, there are no ICRP estimates of percent deposition in these tissues.

Gonads -- Richmond and Thomas reported that in five animal species 0.03% of plutonium was transferred from blood to gonads (29). The MRC review of plutonium toxicity concluded 0.05% was transferred to gonads (18). These observations postdate the ICRP review (20) and should be used.

Excreta -- About 3% of the plutonium is located in urine and feces, primarily in feces (see soft tissues above). There is evidence that higher specific activity isotopes, e.g., ^{237}Pu and ^{238}Pu, are translocated more rapidly than ^{239}Pu. This difference probably reflects the reduced amount of polymeric material in high specific activity radionuclides.

In making estimates of the distribution of transuranics, attention is paid both to the chemical form and particulate size of the element administered. However, little emphasis is given to the question of whether the element was in monomeric or polymeric form. Although this question may influence dose-response estimates, there are little data about the chemical and physical form of transuranics in environmental situations.

In addition, it is known that particles up to 75 μ diameter can pass fairly rapidly and easily into the circulatory system after ingestion by the process of persorption (30). Macrocolloids of polymeric transuranics could be absorbed in a similar manner.

Likewise, inhaled particulates are considered to be transported to the lymphatic system by being transported there by macrophages or penetrating the alveolar membrane physically by endocytosis or pinocytosis (31). The particulates are then carried through the lymphatics to lymph nodes or other sites or dumped into systemic circulation through the thoracic lymph duct. Support for this concept is found in the report that $^{239}PuO_2$ particulates with a mass median diameter of 0.3 μm and a geometric standard deviation of 1.6 were identified in a tracheobronchial lymph node removed from an occupationally exposed worker (32). The particles to which the worker may have been exposed had mass median diameters of 0.28 to 0.45 μm and geometric standard deviations of 1.4 to 1.6 (32). These observations strongly suggest that some particles are transported directly to local lymph nodes after inhalation without changing their characteristics much from those of the inhaled aerosol.

Since macrocolloid or other particles probably reach systemic circulation after inhalation or ingestion of transuranics, it is reasonable to use human health effects data based on other multivalent, colloid formers, i.e., thorium and polonium. This is particularly true when considering the health effects in specific organs such as the liver.

Table I

Disposition of Inhaled Transuranics (25)

Inhaled Class Y Compounds (1.0 μm AMAD)

Region	Percent Deposition (D)	Percent Transported to Blood	GI Tract	Lymph Nodes
Nasopharyngeal	29	0.23	28.71	---
Tracheobronchial	8	0.08	7.92	---
Pulmonary	23	1.15	18.40	3.45
Total	60	1.46	55.03	3.45

Inhaled Class W Compounds (1.0 μm AMAD)

Region	Percent Deposition (D)	Percent Transported to Blood	GI Tract	Lymph Nodes
Nasopharyngeal	29	2.9	26.1	---
Tracheobronchial	8	4.0	4.0	---
Pulmonary	23	3.45	18.4	1.15
Total	60	10.35	48.5	1.15

Ingestion of Transuranics

Absorption of transuranics from the GI tract has been estimated in the past to be 10^{-4} percent for insoluble forms such as PuO_2 and 3×10^{-3} percent for more soluble forms (20). However, the absorption has been shown to be much greater for strongly acid solutions and chelated forms of plutonium (up to about 2%) and in younger animals (possibly a factor of 100 increase in absorption) (20).

Recent reports suggest that organically bound transuranics are more easily transported across the gut than inorganic forms. More Pu in milk was absorbed than was Pu from Pu-citrate solution (33). This was particularly true in nursing animals which absorbed up to 3.2 percent of ingested Pu in milk, 1000 times the ICRP estimate of the absorbed fraction (33). In studies at Battelle Northwest Laboratory, biologically incorporated (protein bound) Pu was absorbed up to 10 times as readily as Pu in Pu-nitrate, while absorption of similarly prepared Np was only about 1/10 to 1/20 of that seen with Np-nitrate (34).

Earlier, Sullivan and Crosby (35) had shown that rats given single oral doses of transuranic isotope absorbed from 0.1 percent to 0.01 percent of nitrates and oxides in the case of adults and 0.5 percent to 5 percent of nitrates and oxides (except Pu oxides) in the case of neonates.

At the request of the Agency, scientists at Battelle-Pacific Northwest Laboratory estimated the absorption of transuranic elements from the gastrointestinal tract. They concluded that for oral administration a prudent estimate for radiation protection purposes would be that persons one year of age and older would absorb: 0.1 percent of all inorganic transuranics except ^{239}Pu and ^{240}Pu oxides; 0.01 percent of ^{239}Pu and ^{240}Pu oxides; and 0.5 percent of biologically incorporated transuranics (36). Infants (less than one year of age) would absorb 10 percent of all transuranic elements except ^{239}Pu and ^{240}Pu oxides; and 1 percent of ^{239}Pu and ^{240}Pu oxides (36).

While these estimates have a large degree of conservatism and are more than an order of magnitude greater than current ICRP estimates, they are supported by other observations. In particular, a review of gonadal deposition of actinide elements reported that, following intravenous injection of plutonium (citrate or nitrate), gonadal deposition in the pig was about 2×10^{-4}; following ingestion about 3×10^{-6} (29). This suggests absorption of a fraction about 10^{-2} of the amount ingested. In the case of beagles, the fraction of plutonium deposited in the gonads following intravenous injection was about 4×10^{-4}, following ingestion about 8×10^{-6} (29), again indicating about 10^{-2} is transferred through the gut wall. These observations support use of the Battelle estimates rather than those of ICRP.

The current ingestion model used by ORP is patterned after the four compartment model of ICRP-2 (37), but neglects the one-hour time delay in the gastrointestinal tract. This model is combined with the lung model and used both for the ingestion pathway and as part of the inhalation pathway for those lung subcompartments clearing through the gastrointestinal tract (26).

Skin Contamination

The case of skin contamination as a route of exposure for transuranics can be neglected unless the skin is not intact. Even if the skin is damaged, there are not enough data to make a quantitative estimate of the rate of amount of absorption of environmental transuranics. However, this mode of exposure is believed at present to be a relatively unimportant pathway for the case of interest here.

PROBLEMS IN DOSIMETRIC MODELING FOR TRANSURANIC ELEMENTS INHALATION

Inhalation

The physical, physiological, and biological assumptions used in the models for dosimetry and metabolism used in toxicologic studies of the transuranics have some problem areas.

To some extent the ICRP Task Group on Lung Dynamics (TGLD) model (25) employs parameters that maximize dose estimates in its assumptions for exposure by inhalation. To this extent it is conservative. The model assumes tidal volumes of 750 cc or 1450 cc for resting and light work, respectively, which are adequate for an adult male. Values for adult females are of the order of 340 cc or 660 cc, and for children they drop as low as 17 cc or 33 cc in the sleeping newborn (27,38). In this respect, the model provides estimates of exposure which are somewhat higher than would be expected in a general population. This is compensated for in part by the decreased organ mass for children and women which leads to higher average organ dose.

The TGLD model uses other assumptions which are not conservative:

1. The model uses a respiratory rate of 15 respirations per minute. For a heterogenous aerosol the percentage of deposition varies with breathing rate. The minimum level of deposition occurs at 15 to 20 respirations per minute and increases on either side of this value (39). Deposition rates in hard workers, or in sleeping or sedentary individuals would be higher than the model predicts.

2. The Task Group, as Mercer points out, used a Findeisen anatomical lung model, which underestimates airway branching and therefore impaction sites (40). This will lead to overestimation of pulmonary deposition and underestimation of tracheobronchial deposition during mouth breathing (40).

3. The pulmonary lung is not treated as a single compartment but rather as a series of parallel tubes so that clearance by a given pathway is arbitrarily independent of other clearance routes.

4. No provision is made in the model for the known regional distribution of inhaled material within the lung. Bates, Ball and Bryan have shown that, in the upright individual, the distribution and rates of wash-in and wash-out gases are different by about 40% between the upper and lower lobes of the lung, indicating a similar difference in regional airflow (41). This has profound implications in that it affects the distribution of inhaled aerosols both for settling and for diffusion within alveoli.

5. The model is based on laminar flow in tubes at a constant rate, per the calculations of Findeisen and Landahl (25). A summary of the assumptions in the model that need refining include (39):

a. The pattern of airflow during respiration is not constant, but goes from zero to some maximum and then returns to zero. Information is needed on the effect of this pattern on deposition.

b. Airflow in the lung may be a mixture of laminar and turbulent flows. The extent of this phenomenon and its effect on deposition should be investigated.

c. The bulk of new air does not mix volumetrically with lung air. Nondiffusible particles (>0.5μ) will penetrate only as far as the new air goes, while finer particles (<0.5μ) will be able to penetrate the depths of the lung by diffusion similar to that of a gas molecule. This has implications for distribution of alveolar versus respiratory bronchiolar deposition of particles and subsequent clearance by mucus.

d. The respiratory tree is not composed of circular tubes but irregular cross-section tubes which are often corrugated or folded over. The effects of these irregularities on turbulence and deposition are not known.

e. The effects of respiratory excursions (coughs) on respiratory clearance cannot be assessed. This is probably an important route of transport where the mucus escalator is impaired due to smoking, etc.

While the lung model is considered to be accurate enough for standard setting purposes, problems peculiar to plutonium distribution and retention are not settled. There is evidence that the higher specific activity isotopes of plutonium (^{238}Pu, ^{237}Pu) have a different distribution pattern in the body than ^{239}Pu after translocation (42,43). In general, the ^{238}Pu resembles injected monomeric ^{239}Pu* in its distribution pattern after translocation from the lung. When ^{239}Pu is translocated from the lung, the pattern of distribution within the body is similar to that of injected polymeric ^{239}Pu.** This suggests that the ^{238}Pu is dissolving and being transported as a Pu-transferin complex*** within the body while the ^{239}Pu is being engulfed and transported as particulate material (44).

It has been suggested that the more rapid translocation of ^{238}Pu relative to ^{239}Pu aerosols deposited in the lung is due to the effect of the specific activity on local chemistry after deposition; that is, the higher activity ^{238}Pu produced enough radicals in its aqueous environment (lung mucus or parenchyma) to influence local chemistry and the rate of dissolution of the particle (45). Regardless of whether these arguments are correct or not, separate models should be developed for each plutonium isotope based on observed and expected differences.

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*Monomeric plutonium is in the form of single molecules of the plutonium compound, not a large number of particles aggregated.

**Polymeric plutonium is a form where a number of molecules of the plutonium compound aggregate together as colloid.

***Transferin is the serum protein which binds iron as an iron-transferin complex to transport the iron throughout the body. It also appears to be involved in transport of transuranic elements.

The available estimates of half-times for the translocation of plutonium from alveolar deposits and from lymph nodes are subject to considerable uncertainty. The data on retention, particularly in lymph nodes are based on animal experiments at relatively high levels of exposure which impaired local histology and physiologic processes. Therefore, it may not adequately reflect transport in humans exposed environmentally.

Much of the environmentally distributed plutonium is in the form of very small sub-micron particles weakly attached to larger dust particles (46). Unfortunately, the TGLD models does not address the question of extremely small particles bound to 1.0 μ (or larger) particles. This question should be addressed since plutonium which escapes through high efficiency particulate filters is a source of environmental contamination.

Ingestion

The recent studies of transuranium element uptake following ingestion suggest that earlier estimates may be in error. Sullivan and Crosby found (34,35) that 10^{-3} to 10^{-4} of a single ingested dose of transuranics is absorbed from the gastrointestinal tract. This is at variance with the ICRP estimate of 3×10^{-5} to 10^{-6} (20) derived from chronic feeding studies. The estimate of 10^{-3} to 10^{-4} is supported indirectly by data on gonadal deposition of transuranium elements. In the data reviewed by Richmond and Thomas (29) the ratio of gonadal deposition following ingestion of transuranics to that following intravenous injection ranges from about 10^{-2} to 10^{-3}. This implies absorption of about that fraction from the gastrointestinal tract.

The question of the fraction of ingested transuranium element absorbed from the gastrointestinal tract as a function of age of the subject and chemical form ingested should be resolved to a greater extent than it is today. An adequate answer may change dose and risk estimates by an order of magnitude or more.

Distribution and Retention

Dose models currently employed must extrapolate animal data on distribution and retention of transuranium elements to man. While the general lack of data in man demonstrates adequate radiation protection efforts, it does force extensive use of animal data. The animal data usually encompass only one or two ages, one or two routes of administration, and one or two chemical forms of the element in a given sex and species. Often the studies are not directly comparable and it must be assumed that differences in age, sex, or chemical form of the element cause negligible differences in distribution or retention.

As was pointed out earlier (Animal Studies) what data are available on different transuranium elements show that each has its own distribution pattern (13,22). In addition, when sexes or several species can be compared, distribution is often species and isotope specific (22).

Superimposed on these uncertainties in the distribution of transuranium elements is the uncertainty in retention. Most studies have been done in short-lived animal species. However, retention half-times in man are estimated at 40 years or 100 years (20). In a rodent with a two-year lifespan an element with a 40-year half time would decay about 4% and an element with a 100-year half time about 2%. Such variations are less than counting error and individual variation. Even in canines with a 15-year lifespan, decay would be only 16% and 10%, respectively.

It is obvious that more data on man are needed for all actinide elements. Perhaps a solution to the distribution question may come from autopsy series of environmental and occupational exposures and for the retention problem from accident cases or tracer studies. Perhaps additional studies in animals scaled to duplicate specific human experiences for which there are data would strengthen confidence in the extrapolations from animal to man now made so blithely.

Genetics

One of the greatest uncertainties in health effects estimates for transuranium elements relates to the estimate for genetic effects. Not only do the estimates of percent of administered nuclide deposited in gonads from blood have a range of 10^{-1} to 10^{-3} (18,29), but the estimates are almost entirely for males. In addition, what data are available in man from environmental exposure do not agree well with the data from occupational exposure or animal studies (see Annex 3, Section 3.2).

The internal distribution of the transuranium elements in the testes has been reported for mice (47), rats (48), and Chinese hamsters (49). The preferential deposition of plutonium along the peritubular membrane produces an inhomogeneous dose distribution. The inhomogeneity factor was 2.5 for mice, 1.6 for rats and 1.0 for Chinese hamsters (47-49). There is no information on what it might be for man or other species.

There is little published information on inhomogeneity factors for ovaries. There is no good information on distribution from blood to ovary, retention in ovary or testes, nor RBE for alpha emitters in the ovary or testes (although some recent work suggests an RBE for testes of 20 or more).

The uncertainty in genetic risks from exposure to transuranium elements is quite high, probably orders of magnitude. Studies to identify retention, distribution, inhomogeneity of dose, and cytogenetic effects adequately are urgently needed to determine if somatic or genetic effects constitute the primary hazard of exposure to transuranium elements.

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ACUTE TOXICITY OF
TRANSURANIUM ELEMENTS

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ACUTE TOXICITY OF TRANSURANIC ELEMENTS

Although plutonium and transuranics are reported to be extremely radiotoxic, the magnitude of toxicity is closely related to the route of administration. There is no evidence that transuranics can compare with highly toxic chemical or biological agents when the route of administration is by injection (1,2). Following ingestion or inhalation, the toxicity of transuranic elements is probably comparable to that of the most toxic materials known.

The first consideration of transuranic element toxicity should be for acute responses following relatively massive exposures to the material. Such responses may be characterized as peracute, acute, or subacute.

Peracute Toxicity

For purposes of discussion peracute toxicity will refer to the case when the endpoint, death, occurs within hours of exposure.

1. Inhalation

There is no substantive evidence that inhalation of transuranic elements will cause death in hours due to radiotoxicity. Undoubtedly, air concentrations of transuranic element aerosols sufficiently high to cause death by smothering can be estimated. However, the same concentration should then be effective for even an inert aerosol. Inhalation of many chemicals and biologic agents in the mg/kg range or less will kill in hours (1,2,3,4).

2. Ingestion

Tentative evidence of peracute toxicity associated with massive oral doses of plutonium was reported by Sullivan and Thompson in 1957 (5). They found that 50% of rats given an oral dose of 93 mCi/kg of plutonium nitrate died within one day. The dose would be about 18 mCi per rat, equivalent to 294 mg of ^{239}Pu or 600 mg of $^{239}Pu(NO_3)_4$. If this dose does cause peracute toxicity, and is scaled up to a 70-kg man on the basis of body weight, the oral doses associated with peracute radiation lethality following ingestion by man would be: ^{239}Pu - 106 grams, ^{238}Pu - 375 mg, ^{241}Am - 2.45 grams, and ^{244}Cm - 95 mg. If the mass of the gastrointestinal tract rather than the body weight was used in developing the scaling factors, the respective oral dose estimates would be: ^{239}Pu - 48 grams, ^{238}Pu - 170 mg, ^{241}Am - 1.11 grams, and ^{244}Cm - 43 mg. Many chemical and some biological agents when administered orally are fatal at levels of μg/kg (1,3,4).

Sullivan and Thompson (5) doubted that this peracute response of rats to plutonium nitrate was due to radiation injury. Sullivan, et al. (6), showed that an ingested beta emitter, Ru-106, could cause acute lethality. However, they did not see any deaths in less than four days after exposure. The possibility of acute nitrate toxicity has also been considered, but the question of peracute nitrate toxicity is complicated by the differences in the blood enzyme methemoglobin reductase between species and ages. The young adult appears to have the highest reductase activity, adults have less, and very young animals only limited activity (7). Sheep reduce methemoglobin more rapidly than pigs or horses (7).

The only specific estimates of nitrate toxicity in rats appear to be those of Wright and Davidson (8) who found that the LD_{50} for intravenous injection of nitrate is 3.452 g/kg (800 mg/kg of nitrate-nitrogen); and Druckrey, et al. (9), who reported that 443 mg/kg (100 mg/kg of nitrite-nitrogen) of nitrite fed to rats in drinking water caused growth inhibition and shortening of lifespan.

On this basis, the 300 mg of nitrate in the 600 mg of plutonium nitrate given to each rat would not be expected to be the cause of peracute death reported by Sullivan and Thompson (5). Until some better evidence is available, the cause of death cannot be attributed to nitrate toxicity.

The toxicology of nitrates is confused by the toxicology of nitrites. In the production of methemoglobinemia, conversion of hemoglobin in red blood cells to methemoglobin which cannot transport oxygen in the blood, the controlling factor appears to be the rate of conversion of nitrate to nitrite since nitrite seems to be the agent causing the methemoglobinemia (7,10,11). While anoxia develops as a consequence of methemoglobinemia, the exact mechanism of cause of death is not known (12). Normally 1 to 2% of total blood hemoglobin is methemoglobin (7). Methemoglobin concentrations of 30 to 50% lead to anoxic symptoms but are compatible with life (7,12). Death occurs at methemoglobin concentrations of 50 to 80% of total blood hemoglobin (7,12).

Conversion of 10% of hemoglobin to methemoglobin is estimated to be caused by 1 mg/kg of nitrite and 2 mg/kg of nitrate in infants less than three months of age. A 10% concentration of methemoglobin in the blood, considered the upper limit of the subclinical range, is generally regarded as of no medical importance (7). Burden (13) estimated the maximum permissible dose of nitrate as 53 mg (12 mg nitrate-nitrogen) in a 3 kg infant and 1.062 g (240 mg nitrate-nitrogen) in a 60 kg adult. Above these levels appreciable methemoglobinemia may occur.

The lethal dose of nitrite has been estimated at 20 mg/kg in the adult (10) and of nitrate at about 120 mg to 600 mg per kg in the adult (13). Acute methemoglobinemia with cyanosis, vomiting, abdominal pain, etc., has been observed in children after sodium nitrate doses of 100 to 400 mg and in adults after doses in excess of 150 mg (11). Peracute death, in minutes, has been observed after doses of nitrite exceeding 1 gram. The death appears to be due to cardiovascular collapse and shock (11). So it is probable that peracute transuranic nitrate radiation death could not occur in man; peracute nitrate toxicity would probably occur first. However, since the peracute lethality following massive oral doses of plutonium nitrate in the rat is not due to nitrate toxicity, the possiblity remains that there is a transuranic element toxicity which is as yet unidentified. While radiotoxicity is a possibility, so is a direct heavy metal toxicity.

Acute Toxicity

For purposes of discussion, acute toxicity will refer to the case when the endpoint, death, occurs in days, usually less than 60 days, post exposure. If 50 percent of those exposed die in 30 days, the associated exposure is called the $LD_{50/30}$. If 50 percent die in 60 days, the associated exposure is called the $LD_{50/60}$.

1. Inhalation

Thompson (14) estimated that with an $LD_{50/30}$ for ^{239}Pu-citrate; in rats of 70 μCi/kg, in mice of 70 μCi/kg and in dogs of 20 μCi/kg following intravenous injection, a minimum estimate of the internally deposited dose which would be an $LD_{50/30}$ was 10 μCi/kg. Based on this $LD_{50/30}$ internal deposit of 10 μCi/kg in animals, the $LD_{50/30}$ for a 70 kg man would be 700 μCi, i.e., 10 mg of ^{239}Pu or 40 μg of ^{238}Pu. This is roughly 80 mg of ^{239}Pu or 230 μg of ^{238}Pu inhaled.

Since curium and americum require a dose about 22% greater than ^{239}Pu to cause 50% lethality in 30 days (15), the approximate doses in man which might produce an $LD_{50\ 30}$ are: ^{241}Am or ^{244}Cm, 854 μCi deposited or about 264 μg of ^{241}Am and 10 μg of ^{244}Cm. This would be roughly 2.1 mg of ^{241}Am or 80 μg of ^{244}Cm inhaled, if the same scaling used for plutonium is applied.

If the estimated dose is scaled according to the amount of isotope per gram of lung, the dose required to produce an $LD_{50/30}$ would be greater. Using the data from Durbin (15), Thompson (16) and Wacholz (1) the estimated doses for an $LD_{50/30}$ are 11.25 to 19.44 μCi/gram of lung in the rat and 10 μCi/gram of lung in the dog. If 10 μCi/gram of lung is used as the $LD_{50/30}$ estimate, the dose required in man is 1000 μCi. This is equivalent to about 14 mg ^{239}Pu, 60 μg ^{238}Pu, 380 μg ^{241}Am or 14 μg ^{244}Cm deposited in the lung; 112 mg ^{239}Pu, 480 μg ^{238}Pu, 2.04 mg ^{241}Am or 112 μg of ^{244}Cm inhaled.

While these doses are lethal to 50% of the exposed animals in 30 days, curves shown by Buldakov, et al. (17), indicate that early mortality following an $LD_{50/30}$ dose of plutonium may start within five days after the exposure in rats.

2. Ingestion

Acute ingestion toxicity has not been reported as a radiation response following ingestion of transuranium elements. Buldakov, et al. (17), did observe 50% mortality in 99 days in rats fed 50 μCi/kg of ^{239}Pu-citrate per day. Acute ingestion toxicity in a 70-kg man would, on this basis, require daily doses in excess of 50 mg ^{239}Pu, 0.2 mg ^{238}Pu 1.1 mg ^{241}Am, or 41 μg ^{244}Cm; with perhaps half that much required if the mass of the gastrointestinal tract is used rather than total body mass.

Subacute Toxicity

For purposes of discussion, subacute toxicity will refer to the case when the endpoint, death, occurs more than 60 days but less than many years post exposure.

1. Inhalation

Large doses of inhaled transuranics will probably cause fatalities in weeks to a year or so past exposure. The major cause of death will probably be radiation induced pneumonitis and pulmonary fibrosis. Baeza, et al. (18), in a small number of patients, found about 25% developed radiation pneumonitis after x-ray doses of 1500 to 2000 rads given across a two-week period. This will be used as a lower estimate of the radiation dose that will cause death due to radiation pneumonitis and fibrosis.

In animal experiments with inhaled $^{239}PuO_2$ both rats and dogs could be killed by pulmonary fibrosis (19).

Species	Total Lung Dose (rads)	Dose Administered Lung	Whole Body
Rat	20,000	0.7 μCi/g	3.0 μCi/kg
Dog	1600-14000	0.3-0.2 μCi/g	0.2-1.4 μCi/kg

Based on these data, the subacute toxicity for inhaled transuranics in man should start at 30 μCi lung dose. This would be about 490 μg ^{239}Pu, 1.7 μg ^{238}Pu, 9.2 μg ^{241}Am or 0.4 μg ^{244}Cm.

2. Ingestion

Subacute lethality following ingestion has not been reported. Any effects anticipated would be delayed effects (cancers).

CONCLUSIONS

The whole question of peracute, acute and subacute effects of transuranics in man as outlined above is highly speculative. The extrapolations from animals to man may or may not have validity because of other competing factors including mass transport phenomena. However, since human data are not, and hopefully will not become, available, extrapolation of animal data is the best method to estimate the order of magnitude of human risk.

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INHALATION AND INGESTION MODELS FOR HUMANS EXPOSED TO RADIOACTIVE METERIALS

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INHALATION AND INGESTION MODELS

INTRODUCTION

The two primary modes leading to internal radiation exposure are the inhalation and ingestion of radioactive materials. The estimation of organ burden and exposure, as well as of the resulting dose rates and doses, due to uptake by these pathways, is relatively complicated and requires the adoption of mathematical models which depend on many parameters.

A. Inhalation

Industrial hygienists have recognized for many years that the inhalation of an aerosol carrying radioactive nuclides was a potential mechanism for damage to the respiratory tract as well as a possible pathway for the translocation of inhaled radioactive material to other internal organs. The complexity of the biological phenomena which govern transmission and elimination of such material makes consideration of potential health effects due to the inhalation of radioactive materials extremely complicated. Even a first order analysis of the process must consider the factors enumerated below:

1. The fractional deposition of inhaled material in the respiratory tract depends on properties of the aerosol - size and mass distribution, chemical form and charge - as well as on the physiological characteristics - surface properties, configuration, and breathing rate - of the lung.

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2. The duration and extent of the exposure* depends on the biological and physical mechanisms which transport the deposited material within the body. These include the various clearance paths, the nuclide half-life, the chemical form, solubility, and the degree of retention in each organ of interest.

3. The dose depends on the time integral of the activity in the organ, the organ mass, the emitted energy, and the fraction of the energy absorbed by the organ tissues. For alpha emitters, this absorbed fraction is assumed to be unity. At present, the organ mass, breathing rate, and clearance times in the PAID code (see below) correspond to a 30 year old working male. Specific parameters are given in the text.

In some cases, H, the dose equivalent in rems can be found by multiplying the dose (rads) by quality and modifying factors as defined in ICRU supplement 19. In the case of lung tissue, a modifying factor has yet to be established for particulate sources of alpha radiation. Therefore, provisionally, 1 rad (α, lung) is equivalent to 10 rem. In actual practice the risk can be calculated in terms of rad (α, lung) and the use of the dose equivalent becomes irrelevant.

4. The health effects depend on type of radiation, site of energy deposition, susceptibility of the organ to radiation damage, and specific type of health effect considered. Since the mechanisms

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*The time integral of the activity is given the name exposure in ICRP Report #10.

by which somatic and genetic damage are inflicted are incompletely understood, accurate stipulation of the degree and number of health effects is also subject to unquantifiable uncertainties.

B. Ingestion

The ingestion of radioactive material represents another pathway for internal radiation exposure. While description of this pathway is generally considered to be simpler than for inhalation, due to the direct deposition of all the ingested material into the gastrointestinal tract, treatment of the balance of the biological-physical processes involved suffers from many of the same limitations discussed above for the inhalation mode.

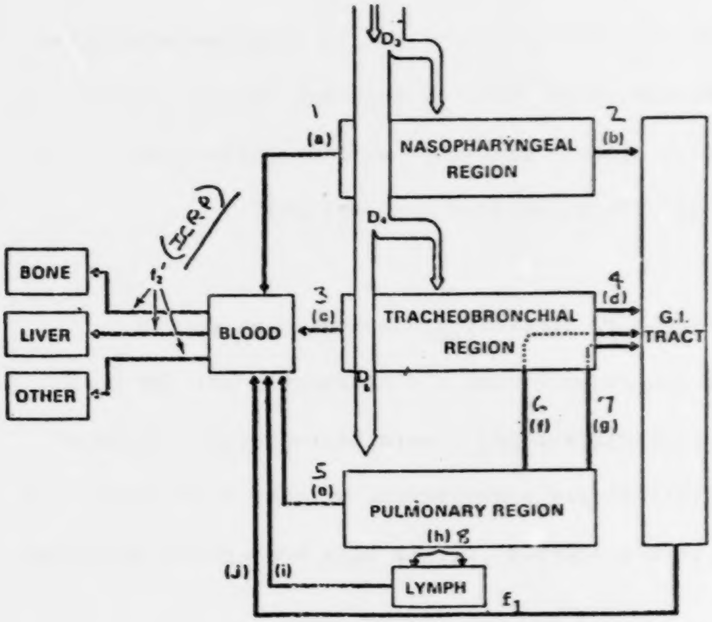
For ingestion, the critical transfer mechanism appears to be the absorption of radioactive materials into the systemic blood. Values for this fraction have been studied in animals and, to a very limited extent, in man but are still subject to large uncertainties which strongly affect projected doses to internal organs. As a consequence, the health effects predicted will be subject to uncertainties until more detailed data are available.

C. Models

Reasonable estimates of internal radiation doses due to inhalation and ingestion require that a consistent model for both the respiratory and gastrointestinal tracts be employed. While a large amount of theoretical and experimental work on such models has been done, the most widely accepted models have been those developed by members of the respective International Commission on Radiological Protection (ICRP) task groups.

The ICRP Task Group on Lung Dynamics (TGLD) has proposed a model for the respiratory tract which has been well documented and the parameters suggested for use in the model have been extensively reviewed and, to some extent, improved in later ICRP publications (Morrow, 1966), (ICRP, 1972). Details of this model are given by Morrow with the revised parameters collected by the ICRP. Therefore, only a brief outline of the model is presented here. The ICRP TGLD proposed model comprises three major compartments: the nasopharyngeal, the tracheobronchial and the pulmonary, as shown in Figure I, taken from ICRP Report #19 (ICRP, 1972).

FIGURE I



Each of these major compartments is divided into subcompartments, corresponding to various transfer mechanisms, which are treated as essentially independent processes. In addition, the associated lymph nodes are appended to the pulmonary compartment in one of the transfer chains. Direct deposition through inhalation is only to the three major compartments with the fractional deposition to each a function of the aerosol properties. Subsequent transfer and/or clearance is governed by the parameters specified for each subcompartment, as shown in Table I (ICRP, 1972).

TABLE I

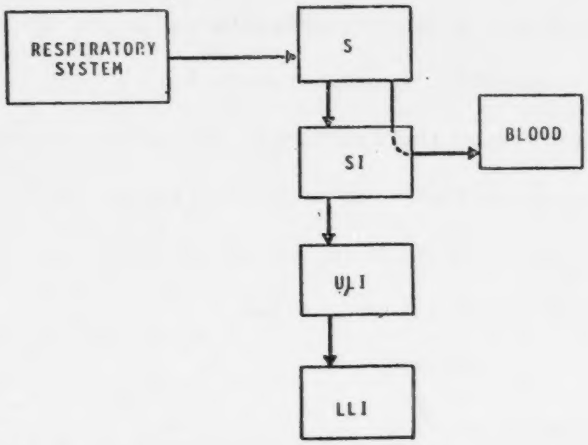
AMENDED CONSTANTS FOR USE WITH TGLM CLEARANCE MODEL

Region	Pathway	Compound class		
		(D)	(W)	(Y)
N–P	(a)	0.01 d/0.5	0.01 d/0.1	0.01 d/0.01
	(b)	0.01 d/0.5	0.4 d/0.9	0.4 d/0.99
T–B	(c)	0.01 d/0.95	0.01 d/0.5	0.01 d/0.01
	(d)	0.2 d/0.05	0.2 d/0.5	0.2 d/0.99
P	(e)	0.5 d/0.8	50 d/0.15	500 d/0.05
	(f)	—	1 d/0.4	1 d/0.4
	(g)	—	50 d/0.4	500 d/0.4
	(h)	0.5 d/0.2	50 d/0.05	500 d/0.15
L	(i)	0.5 d/1.0	50 d/1.0	1000 d/0.9

Ultimate disposition in this model is to the systemic blood or to the gastrointestinal tract.

The ICRP gastrointestinal tract (GIT) model is documented in (ICRP, 1959). The model comprises a four compartment, as shown in Figure II, tract consisting of the stomach, small intestine and lower and upper large intestine. Parameters for this model are given by the ICRP, (ICRP, 1959).

FIGURE II



Although both models are described exhaustively in words, neither of the ICRP groups has given mathematical descriptions for these processes. This lack has led to some confusion in attempting to calculate doses and effects using the models. For the present treatment, several of the previous analyses have been reviewed and discrepancies in the equations compared with the "official" verbal descriptions of the models. While some ambiguities may remain, the present treatment has attempted to reconcile the ICRP descriptions with the governing equations used in the EPA code "Plutonium Air and Ingestion Dose (Sullivan, 1977).

The PAID code calculates the dose from each radionuclide in a two-member chain. Such mother-daughter chains are of particular interest in the dosimetry of transuranium elements because some have a short half-life mother, e.g. curium-242, and others have a beta emitting parent and an alpha emitting daughter, e.g. Pu-241. Complete equations for two-member chain analysis are given in Reference 8. The description here is confined to single radionuclides. This allows a simpler mathematical development. The extension of the LaPlace transform analysis, outlined below, to the chain problem is straightforward.

D. The Respiratory-Gastrointestinal Tract Model

The code used by the Environmental Protection Agency (EPA) corresponds to descriptions of the physiological processes as contemplated by the originating ICRP groups. In addition, an attempt has been made to keep the resulting mathematical relationships as simple and understandable as practicable. To this end, the respiratory and gastrointestinal tract models have been coupled as outlined below:

The ICRP TGLD lung model implicitly assumes that the physiological processes associated with each subcompartment operate independently. Thus, a general equation governing the behavior of any member, either subcompartment or organ, in a chain of members connected in series may be written as:

$$\dot{q}_\ell(t) = S_\ell(t) - \lambda_\ell q_\ell(t) \qquad (1)$$

where q_ℓ = the organ or subcompartment burden in the ℓth organ (curies)

S_ℓ = the source term for the ℓth member (curies)

λ_ℓ = the total, or effective, decay constant for the radionuclide in the ℓth member (yr^{-1})

The source term will vary, depending on the position of the member in the chain. The only direct deposition in the lung is through inhalation and is to k major compartments; nasopharyngeal, tracheobronchial, and pulmonary. These major compartments are further divided into ℓ subcompartments, and their associated pathways, (a) through (h), see Figure 1. For these subcompartments, the source term will be:

$$S_\ell = ID_k f_\ell$$

where

D_k = deposition fraction for the kth compartment

f_ℓ = fraction of D_k translocated through pathway ℓ

and where I may be a quantity (curies) for acute intake or a rate (curies/year) for constant continuous intake, Table II.

TABLE II

Breathing Rate - Male Adult ICRP Report #23 (ICRP, 1975)

	Minute Volume (liters/minute)	Duration (hours/day)
Light Activity	20.0	16
Resting	7.5	8

Average Daily Intake = 2.3×10^4 liters.

For ingestion, the initial compartment is the stomach and the source to this member is:

$$S = I$$

where, again, I may be either the annual intake in curies for continuous ingestion, or a quantity (Ci) for an acute intake.

For succeeding members of the chain, the only source is material transmitted from the preceeding organ. All organs or subcompartments inferior in position are presumed to have no initial material content. Therefore, for all subsequent organs

$$S_\ell(t) = \lambda^b_{\ell-1}\, q_{\ell-1}(t) \qquad (2)$$

where ℓ-1 refers to the preceeding member and λ^b is the biological decay constant. It is obvious, from the last equation, that each equation in the chain is coupled to all preceeding equations through the source term.

Coupled equations of this type are most readily solved by using LaPlace transforms. Application of the transform to Equation 1 yields

$$sq_\ell(s)-q_\ell(o) = S_\ell(s)- \lambda_\ell q_\ell(s)$$

whence

$$q_\ell(s) = \frac{S_\ell(s) + q^o_\ell}{(s + \lambda_\ell)} \qquad (3)$$

and q^o_ℓ is the initial burden (Churchill, 1944). The initial organ burden for all organs inferior to the first, $\ell>1$, is set equal to zero. Furthermore, the source terms for the first organ (or subcompartment) are constant and their transforms will be:

$$S_1(s) = \frac{ID_k f_\ell}{s}$$

or

$$S_1(s) = \frac{I}{s}$$

To simplify the notation involved, let the constant part of both of these be represented by I' and let any initial acute deposition, presumed to be present at t = 0, be represented by I_o. The general equation then becomes, in transform space,

$$q_1(s) = \frac{I'}{s(s + \lambda_1)} + \frac{I'_o}{(s + \lambda_1)} \tag{4}$$

for the first member of the chain. Using the transform of Equation 4 as the source in Equation 3, the equation for the second member of the chain is seen to be

$$q_2(s) = \frac{f_2\lambda_1^b\, I'}{s(s + \lambda_1)(s + \lambda_2)} + \frac{f_2\lambda_1^b\, I'_o}{(s + \lambda_1)(s + \lambda_2)} \tag{5}$$

from which it is obvious that the equation for each succeeding member in the same series differs only by an additional factor $(s + \lambda)$ and, of course, by modification of the coefficient by the terms f_ℓ and by $\lambda_{\ell-1}^b$, the transmission fraction to the second organ and the biological decay constant of the first, respectively. The general equation for the nth organ or subcompartment may then be written as:

$$q_n(s) = [\prod_{\ell=1}^{n} f_\ell\, \lambda_{\ell-1}^b] \left\{ \frac{I'}{s[\prod_{\ell=1}^{n} (s + \lambda_\ell)]} + \frac{I'_o}{[\prod_{\ell=1}^{n} (s + \lambda_\ell)]} \right\} \tag{6}$$

where λ_o is defined as 1 and the first term corresponds to the continuous intake case and the last to the acute case. It is easily noted that these terms differ only by the factor s in the denominator of the first. A well-known theorem in transform theory states that division of the transformed equation by s corresponds to integration of the inverse transform between the limits 0 and t (Churchill, 1944). Therefore, the solution to the continuous intake case is the integral of the acute intake case. The time integral of the organ burden equation is defined as the "exposure", $Q_n(T)$, in ICRP, 1969.

$$Q_n(T) = \int_0^T q_n(t)dt = L^{-1} \{\frac{q_n(s)}{s}\} \qquad (7)$$

The general equation for the exposure, in transform space, to any organ is obtained by dividing both terms of Equation 6 by s.

$$Q_n(s) = [\prod_{\ell=1}^{n} f_\ell \lambda^b_{\ell-1}] \{\frac{I'}{s^2[\Pi(s + \lambda_\ell)]} + \frac{I'_o}{s[\Pi(s + \lambda_\ell)]}\} \qquad (8)$$

Inverse transforms for equations of this form are readily found using an extension of the Heaviside partial fraction expansion (Churchill, 1944). Applying this expansion to the first term of Equation 6 yields a general solution for the nth organ burden in the continuous intake case:

$$q_n(t) = \{\frac{K_1}{\prod_{j=1}^{n}(\lambda_j)} + \sum_{i=1}^{n} \frac{K_1 e^{-\lambda_i t}}{-\lambda_i [\prod_{\substack{j=1 \\ j\neq i}}^{n} (-\lambda_i + \lambda_j)]}\} \qquad (9)$$

where $K_1 = \prod_{i=1}^{n} f_i \lambda^b_{i-1}$

and λ^o is defined as 1.

For the acute case, corresponding to the second term of Equation 6, the removal of the s in the denominator is equivalent to differentiation with respect to t (Churchill, 1944). Performing this differentiation on Equation 9 yields the solution for the organ burden for acute intake which is identical to Equation 9 if the first term is omitted and K_1 is replaced by $K_1 \rightarrow -\lambda_i I'_o K_1/I'$. Inverse transforms for equations containing repeated linear factors, while somewhat more tedious to calculate, are still straightforward (Churchill, 1944). The general solution to the first term of Equation 8 for the exposure is

$$Q_n(T) = \left\{ \frac{K_1 T}{\Pi(\lambda_j)} + \Sigma \frac{K_1 e^{-\lambda_i T}}{-\lambda_i^2 \, [\Pi(-\lambda_i + \lambda_j)]} \right\} \qquad (10)$$

where the indices run as in Equation 9. This solution may perhaps, be more readily verified by conventional integration of the burden solution, Equation 9. The solution for the exposure due to acute intake then follows from the treatment outlined for the acute burden but, as described above, is identical to Equation 9. For the lung, the average dose is defined here as the average dose to the pulmonary compartment (with a mass of 570 grams) which, for Class Y compounds, receives the greatest dose. This dose can be used to determine an upper bound on the risk of lung cancer. The dose rate and dose are defined in terms of the organ (or subcompartment) burden and exposure as:

$$\dot{D}(t) = 51.2 \frac{\varepsilon}{M_n} q_n(t) \qquad (11)$$

and

$$D(T) = 51.2 \frac{\varepsilon}{M_n} Q_n(T) \qquad (12)$$

where

ε = the absorbed energy (MeV) per disintegration for a particular isotope and organ pair.

m_n = the mass of the organ (grams)

and the result is in rads (ICRP, 1968).

Finally, the health effects are estimated by multiplying the dose for each organ by the number of effects expected per unit dose.

E. Solutions for a Single Radionuclide

Due to similarities in each of the terms of Equations 9 and 10, the solution for each succeeding organ may be found by iterating on the previous solution. For example, consider the chain consisting of the lung subcompartment, h, which transfers material to the lymph nodes, i, and, thence, through the systemic blood, to a reference organ, n. From Equation 9, the solution for the burden in the h subcompartment, for the acute intake case, is:

$$q_h(t) = ID_5 f_h [e^{-\lambda_h t}]$$

Representing the terms in this solution by H^1 and H^2, the solution for the lymph node is:

$$q_i(t) = f_i \lambda_h^b H^1 \left[\frac{H^2}{(-\lambda_h + \lambda_i)} + \frac{e^{-\lambda_i t}}{(-\lambda_i + \lambda_h)}\right]$$

and, using L for those terms, the solution for organ n is:

$$q_n(t) = f_n \lambda_i^b \; L^1 \left[\frac{L^2}{(-\lambda_h + \lambda_n)} + \frac{L^3}{(-\lambda_i + \lambda_n)} + \frac{e^{-\lambda_n t}}{(-\lambda_n + \lambda_h)(-\lambda_n + \lambda_i)}\right]$$

To simplify the coding as much as possible, the procedure followed is to obtain the solutions for each subcompartment in a chain by starting with the first and modifying succeeding ones as indicated above. When the solutions for all the chains are found, they are then summed to obtain the total doses for each major compartment or organ.

For ingestion, there is only one initial compartment, the stomach, and, therefore, only one chain. Since, in the ICRP model of the gastrointestinal tract, the only transfer of material to the blood is accomplished in the small intestine, only the stomach and small intestine ICRP compartments are built into the program. If results for the upper large intestine (ULI) and the lower large intestine (LLI) are desired, they must be run as reference (user supplied) organs.

Two additional modifiers for the equations must also be considered. First, for material which is transferred through the gastrointestinal tract, the additional fraction f_1 (transfer from small intestine to blood) must be used as multiplier for organs inferior to the small intestine. Second, for material transferred through the systemic blood, the fraction f_2' (fraction from blood to reference organ) must be incorporated into the product of the transmission fractions. These parameters are automatically inserted into the chains by the program.

Solutions for the mother-daughter chains follow the same form as those shown alone, but are of course more complex due to consideration of the multiplicity of sources for the daughter radionuclide. The daughter may be formed by decay of the mother radionuclide in a given compartment or may enter that compartment due to decay in and subsequent transfer from all previous compartments. The solutions are complex, see Reference 8.

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EVALUATION OF
SAMPLE COLLECTION AND ANALYSIS TECHNIQUES
FOR ENVIRONMENTAL PLUTONIUM

David E. Bernhardt

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PREFACE

The Office of Radiation Programs of the U.S. Environmental Protection Agency carries out a national program designed to evaluate population exposure to ionizing and non-ionizing radiation, and to promote development of controls necessary to protect the public health and safety. This literature survey was undertaken to assess the available information concerning sampling and analysis techniques for environmental concentrations of plutonium. Readers of this report are encouraged to inform the Office of Radiation Programs of any omissions or errors. Comments or requests for further information are also invited.

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Director, Office of
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This report has been reviewed by the Office of Radiation Programs - Las Vegas Facility, Environmental Protection Agency, and approved for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

EVALUATION OF SAMPLE COLLECTION AND ANALYSIS TECHNIQUES FOR ENVIRONMENTAL PLUTONIUM

ABSTRACT

Information concerning sampling and analysis techniques for plutonium in the environment is presented and evaluated in this report. Consideration is given to available techniques and their applicability to various situations, sensitivities of the techniques, and the validity and reproducibility of results.

Soil is the primary reservoir for plutonium in the environment but inhalation, with the resulting lung dose, is the primary pathway for human exposure. This evaluation is therefore primarily oriented toward sampling and analysis of soil and air, with secondary consideration of other environmental media.

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The author, although recognizing the assistance of many people, accepts full responsibility for the content of this report.

INTRODUCTION

OBJECTIVE

The objective of this report is to review and evaluate past, present, and proposed environmental sample collection and analysis techniques for the measurement of plutonium and associated transuranic elements. Consideration is given to the various available techniques, their applicability to various situations, sensitivities of the techniques, and reproducibility of results.

Soil sampling appears to be the predominant technique for assessing accumulative environmental levels of plutonium (AEC, 1974). Thus, emphasis in this review has been placed on soil sampling and analysis, although consideration is given to other media, especially air sampling. Air sampling is emphasized because of the predominance of the inhalation pathway for plutonium. The review is largely based on published information from nationally recognized laboratories, although some unpublished data, which may include unintentional bias, is included.

GENERAL STATUS OF TECHNIQUES AND THEIR EVALUATION

There are several published intralaboratory evaluations of analytical techniques (e.g. Chu, 1971; Bishop et al., 1971; Sill, 1971; Sill and Hindman, 1974). There are also several reports containing limited data from interlaboratory comparisons (Krey and Hardy, 1970; AEC, 1973; Sill and Hindman, 1974). These studies have largely dealt solely with analytical techniques for soil samples, with limited consideration of the interaction between sample collection and analytical techniques. Krey and Hardy (1970) and Bliss (1973) present some data on the interaction of both collection and analysis, but there does not appear to be any comprehensive evaluation of both collection and analytical techniques.

Most analytical cross-check programs intra- or interlaboratory are done with samples containing plutonium concentrations significantly above background (roughly 0.05 pCi/g of dry soil for a 5 cm depth sample). But, there are several limited groups of data available for replicate analyses of samples containing near-background plutonium levels. These are reported by Sill (1971), AEC (1973), Krey and Hardy (1970), and Butler et al. (1971).

Much of the difficulty with the sampling and analysis of soil samples appears to relate to the discrete particulate nature of plutonium contamination under some circumstances. The potential refractory nature of plutonium, along with the potential for producing refractory material during sample preparation aggravates the inherent difficulties and complexities of the analysis (Sill, 1971; and Sill and Hindman, 1974).

Although there are considerable variations and potential inadequacies in past techniques, and to a lesser extent in current techniques, there is cause for optimism for improvements, or at least standardization, in the near future. In May 1974, the Atomic Energy Commission (now Nuclear Regulatory Commission-NRC) issued Regulatory Guide 4.5, "Measurements of Radionuclides in the Environment, Sampling and Analysis of Plutonium in Soil" (AEC, 1974). This Guide outlined generally compatible and supplementary collection and analysis techniques. In April 1974, the Environmental Protection Agency (EPA), National Environmental Research Center-Las Vegas (NERC-LV; now known as Environmental Monitoring and Support Laboratory, EMSL) sponsored a workshop on soil collection and analysis techniques. A summary of this workshop (attached as Appendix A) and the tentative reference method developed from it (Bretthauer et al., 1975) were issued in 1975.

The following paragraphs are extracted from the EPA criteria for standard methods (EPA, 1973).

"Sampling is the removal from the environmental continuum of a portion of the pollutant for detailed investigation. Sampling involves containerizing a discrete volume of polluted air, water, soil, or biological materials, or it may involve partitioning the pollutant directly from these media into a filtering or absorbing device or into another fluid (e.g., the absorption of the sulfur dioxide pollutant in air into a solution of potassium tetrachloromercurate). Additionally, it includes those procedures necessary to preserve the sample. In all of these sampling methods, we must accurately know what fraction of the pollutant passes from the environmental continuum into the sample. Standardization of the sampling method establishes the reproducibility of this relationship. This relationship must be shown to be stable or to follow predictable changes from the time the sample is taken to the time the sample is worked up for analysis."

"Sample work-up consists of the preparation of the sample by concentration of pollutant, removal of interfering substances, etc., for the analytical procedures to follow. It must be established that all pollutant losses during sample work-up can be quantitatively accounted for and are reproducible within statistically acceptable limits."

"Analytical methods are designed to give accurate estimates of the true amount of pollutant remaining in the worked-up sample. The standardization procedure assures that these values are reproducible within statistically-acceptable limits. The value derived from the analytical method adjusted for predictable losses in sampling and sample work-up gives the estimation of the true concentration of the pollutant in the environmental continuum."

"The reference method is the best, readily available method. Under most circumstances, it will be expected that the reference method will be the method of choice of most user laboratories. When other methods must be used for any reason, their equivalence to the performance characteristics of the reference method must be demonstrated to assure that data generated by their use is equivalent to that generated by the reference method and that statistically valid comparisons can be made between such data and that generated by use of the reference method."

EPA started a standards distribution program for plutonium-239 and americium-241 in December 1973 (EPA, 1974a). A plutonium-239 cross-check program for water samples (<10 pCi/l) was initiated in 1974 (EPA, 1974b).

A basic problem in most environmental monitoring programs is inadequate coordination of the sampling and analytical programs. This is exemplified by a field program where significant efforts are made to obtain unbiased soil samples representative of the sampled area. This sample may represent kilograms of material. The analyst, in order to insure complete dissolution of the material, analyzes a one- or possibly ten-gram aliquot of this sample. If the plutonium contaminant is of a discrete particulate nature, replicates from this sample can vary by several orders of magnitude (Bliss, 1973). Therefore, the objectives of the monitoring program must be continually examined and reevaluated.

DIRECT FIELD MEASUREMENT TECHNIQUES

The most viable means of field measurement for plutonium contamination appears to be the FIDLER (Field Instrument for Detecting Low Energy Radiation) instrument developed by the Lawrence Livermore Laboratory (Tinney et al., (1969). The FIDLER uses a thin NaI or CaF crystal (Piltingsrud and Farr (1973)) and photon pulse height discrimination to detect the 17-keV X-rays from the progeny of plutonium, or the 60-keV gamma photon of americium-241. Although the sensitivity of the FIDLER instrument, ideally about 130 nCi/m^2, is about two orders of magnitude above ambient background levels of plutonium (nominally 1-2 nCi/m^2 of plutonium-239, it provides significantly greater utility for contamination surveys than the prior alpha detection survey instruments.

Minimum sensitivities or calibration factors in terms of pCi/m^2 are generally not stated for most alpha survey meters (Dummer, 1958). Survey instruments are generally only designed for assessing the relative degree of contamination. Information from general sources, including Dummer (1958) indicates a general sensitivity, under ideal field conditions, of about 5-10 pCi/cm^2 (50-100 nCi/m^2). The response relationship is about 500 cpm per 100 nCi/m^2. However, these relationships assume that the alpha activity is essentially emitted from an infinitely thin layer of contamination on a smooth surface. Further, the measurement is made with a fragile mylar-windowed probe, which must essentially be placed in contact with the surface. A layer of moisture (dew) essentially will shield out the alpha particles. There are problems of fracturing or contaminating the probe. Vegetation or rocks make it very difficult to place the probe near the surface. Measurements taken at one centimeter from the surface are in error by roughly a factor of two (Dummer, 1958).

Table 1 presents sensitivities and calibration factors for the FIDLER instrument. These values are based on a nominal background of 200 cpm for the 17-keV region and 600 cpm for the 60-keV region. These values assume the background is known within counting error variations. The 17-keV sensitivities relate to a net background counting rate of 75 cpm, above the background of 200 cpm. Thus, an uncertainty in background of 100 cpm, which is possible assuming the background was determined in a distant contamination-free area, introduces a factor of two error.

TABLE 1. SENSITIVITIES AND CALIBRATION FACTORS FOR FIDLER INSTRUMENTS

(1/16" NaI(Tl) crystal)

Nuclide	Energy Region (KeV)	Minimum Sensitivity (nCi/m^2)	Response ($cpm/nCi/m^2$)	Minimum Sensitivity Point Source (nCi)
Plutonium-238	17	56	1.3	28
Plutonium-239	17	130	0.58	63
Americium-241	17	19	3.9	9.4
Americium-241	60	36	36	19
100% photon	17	7	10.1	3.6
100% photon	60	13	100	6.9

Lindeken et al. (1971) studied the background in the 17-to 60-keV energy region. He concluded that although the background may vary by a factor of two in adjacent areas, the energy spectral shape, or the percentage of the background per 10-keV interval, varied by less than 5 percent. Thus, in the absence of general fission product gamma fluxes, the background at about 80 keV (Compton continuum) can be measured within an area of suspected plutonium contamination, and the background in the 17- and 60-keV regions estimated. This technique can be used to supplement or replace other background readings, to minimize the errors associated with variations in background.

Piltingsrud and Farr (1973) report on a modified FIDLER-type instrument using a CaF(Eu) crystal. The modified instrument is amenable for field repair and costs less than the NaI(Tl)-type instrument. A sensitivity value of about twice that for the NaI(Tl) instrument is reported.

Tinney et al. (1969) report field tests for the NaI(Tl)-type FIDLER at the Nevada Test Site. They estimated the actual background to be 400 counts/min, with a corresponding sensitivity of about 300 nCi/m^2. It was noted that although alpha survey instruments indicated a higher count-rate for selected point sources, it was necessary to use the FIDLER to find these sources. Furthermore, this field test indicated that for general contaminated areas, the FIDLER cpm readings were roughly ten times the alpha instrument readings, versus the theoretical ratio of about

0.1. This indicates the poor efficiency of alpha monitoring in the field (actual versus theoretical).

REECO reported (NAEG, 1971) that at NTS, with a depth-dispersed source of plutonium-239, most of the 17-keV X-rays were absorbed in the soil. Even using the americium-241 60-keV photon required a correction factor of three. The use of the FIDLER with a multichannel analyzer readout was also suggested for areas with general fission product contamination.

Gilbert and Eberhardt (1974) present data for plutonium-239: americium-241 ratios based on laboratory analysis for both nuclides, and on plutonium-239 laboratory analysis versus FIDLER estimates for americium-241 from NTS. The data are summarized in Table 2, taken from Gilbert and Eberhardt. The data indicate a change in the plutonium-239:americium-241 ratio by isopleth. The isopleths were relative concentration lines determined by FIDLER surveys. Except for the Clean Slate I and II sites, there is good correlation between the plutonium and americium ratios within the isopleths.

The ratios and correlation of the plutonium-239 to americium-241 60-keV FIDLER readings are also given in Table 2. Although the correlation improves with an increase in plutonium concentration, the correlation indicates there is little direct relationship. Figure 1 presents scatter diagrams of the plutonium-239 versus americium FIDLER data.

Although the FIDLER is an effective instrument for mapping general areas of contamination, its use as an accurate predictor of plutonium concentrations in surface soils appears to be limited, based on the NTS situation. Additonal field evaluations are necessary for a more specific conclusion.

Stuart of EG and G reports (1971) the use of gamma spectroscopy from an aerial platform for measurement of americium-241 in soil.

Due to the disagreement between published values of half-lives, and X-ray and photon yields for plutonium and americium, various values are summarized in Appendix B.

In summary, although the minimum sensitivity for the FIDLER is indicated as 130 nCi/m^2 for plutonium-239, this relates to only 75 cpm above minimum background values of 200 cpm. Given the variability in background with values up to 400 cpm, or more, extreme care has to be exercised to accurately assess net contamination at 200 or even 500 nCi/m^2. Without an accurate knowledge of background, values at these levels would have uncertainties approaching 50-100 percent. The data in Table 2 and Figure 1 indicate that even at 100 dpm/g (roughly 50 pCi/g or 500 nCi/m^2), there is limited correlation between the FIDLER results and plutonium-239 radiochemistry results. Use of the 60 keV gamma

TABLE 2. ESTIMATED CORRELATIONS BETWEEN LABORATORY GAMMA SCANS FOR AM-241 AND PU-239-240, AND BETWEEN FIDLER CPM OF AM-241 AND PU-239-240

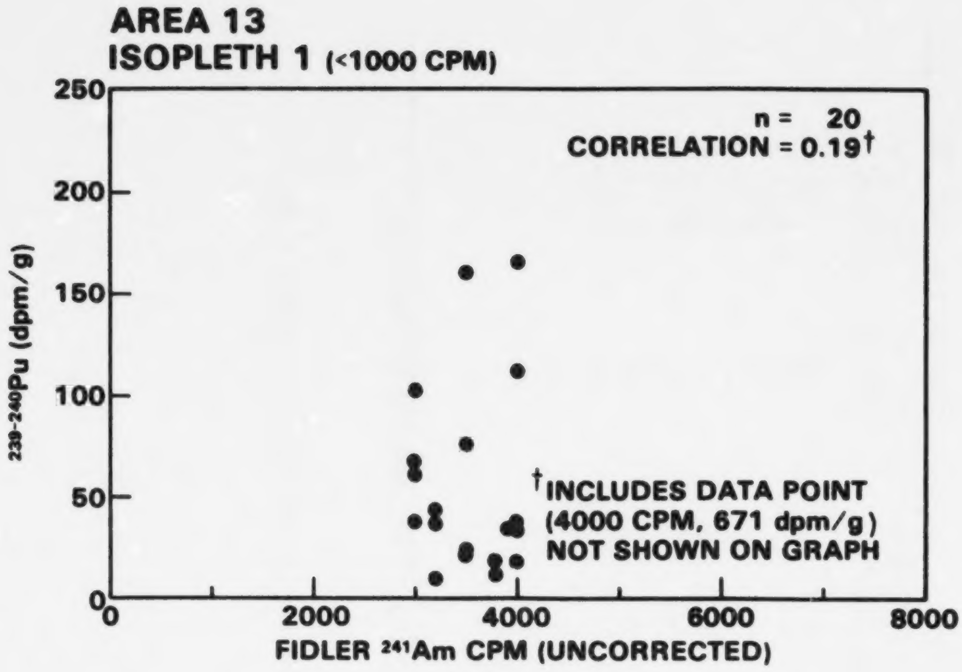
(from Gilbert and Eberhardt, 1974)

		Lab Gamma Scans for Am-241 vs Pu-239-240			FIDLER vs Pu-239-240	
	Isopleth	No. of Samples	Estimated Correlation	Average Pu/Am Ratio ± S.E. †††	No. of Samples	Estimated Correlation
Area 13	1 < 1000 cpm	24	0.98	12.6 ± 0.9	20	0.19†
	2 1-5,000 cpm	28	0.85	14.2 ± 3.9	28	0.33††
	3 5-10,000 cpm	15	0.98	9.4 ± 0.4	14	0.51††
	4 10-25,000 cpm	20	0.99	8.8 ± 0.2	15	0.40††
	5 25-50,000 cpm	20	0.99	8.8 ± 0.3	20	0.69††
	6 > 50,000 cpm	46	0.95	9.4 ± 0.3	46	0.69††
Area 5	1	24	0.93	11.9 ± 1.0	45	0.54†
	5	10	0.99	10.9 ± 0.6	15	0.76†
TTR	Clean Slate I	10	0.73	31.7 ± 5.6	--	----
	Clean Slate II	9	0.54	37.0 ± 10.8	--	----
	Clean Slate III	22	0.91	21.7 ± 2.2	--	----
	Double Track	8	0.99	28.7 ± 1.4	--	----

†FIDLER 60-kev readings not corrected for background (correcting often resulted in negative readings).

††FIDLER 60-kev readings corrected for background.

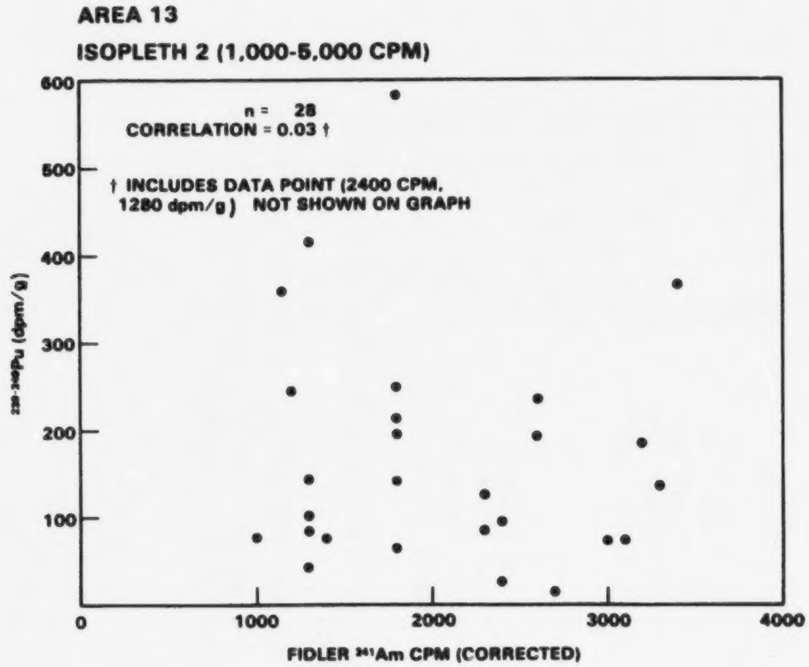
†††These are appropriate only if the Pu/Am ratio remains constant as the Am Value varies. See text for further comments.



AREA 13
ISOPLETH 1 (<1000 CPM)

n = 20
CORRELATION = 0.19†

†INCLUDES DATA POINT (4000 CPM, 671 dpm/g) NOT SHOWN ON GRAPH



AREA 13
ISOPLETH 2 (1,000-5,000 CPM)

n = 28
CORRELATION = 0.03 †

† INCLUDES DATA POINT (2400 CPM, 1280 dpm/g) NOT SHOWN ON GRAPH

Figure 1. Correlation between Plutonium concentrations and FIDLER readings. (from Gilbert and Eberhardt,1974)

from americium-241 for field measurements is not recommended where the age of the material and the original percentage of plutonium-241 is not known.

FIELD COLLECTION TECHNIQUES FOR SOIL

The sampling or program mission and intended use of the sample results is of utmost concern in defining the adequacy of sampling techniques. The efforts and costs associated with sampling as well as the costs of analyzing unnecessary samples make it expedient to relate sampling techniques to the intended use of the data. Table 3 presents approximate costs for sample collection and analysis. The values for collection include nominal driving times between sampling sites.

TABLE 3. APPROXIMATE COSTS FOR SOIL SAMPLE COLLECTION AND ANALYSIS

	Cost	Man-hours per Sample
Sample Collection		
Surface Sample	$10-20	1
Depth Profile (3 to 5 samples)	25-50	2
Plutonium Analysis		
1 gram by Dissolution	100.00	2
10 gram by Dissolution	100.00	4-5
10 gram by Fusion Tech.	150.00	---

There are three primary considerations in sample collection:

1. Selection of the general area to be sampled; e.g., undisturbed, type and amount of vegetation, size of rocks, etc.
2. Determination of sample depth.
3. Compositing material from an adequate sample area.

Appendix A, the summary report from the NERC-LV workshop, includes an extensive discussion of sampling techniques and necessary considerations. AEC (1974) also discusses the criteria.

The intent of sampling programs can generally be related to one or more of five specific objectives:

1. Sampling for low levels, such as those associated with world-wide fallout, to establish base line or background concentrations. The deposition of the contaminant is generally fairly uniform.

2. Sampling to determine the occurrence of a release associated with a specific facility, or accident at a specific location. The deposition levels and distribution may vary with direction and distance from the point of release (Sill, 1971). This includes determining the inventory.

3. Sampling to determine the deposition during various chronological periods of time. The objectives would relate to surface samples or possibly samples from various depths that had been covered at a specific point in time.

4. Profile sampling to determine movement of material through the vertical profile. The sampling technique would be similar to general profile sampling, but samples should not be composited and depths should correspond to the soil horizons.

5. Sampling to determine quantities of source material readily available for resuspension; i.e., normally the surface one-eighth to one-half inch of soil.

Common sampling techniques are not oriented to resuspension. Thus, pertinent comments and techniques are discussed in the next section.

The required accuracy and sensitivity in conjunction with the analytical sensitivity of results must also be considered prior to selecting the sample collection techniques; e.g., dilution of the plutonium concentration in the surface layer by soil with a lower plutonium concentration from deeper profiles. The surface area represented by a sample and the allocation or splitting techniques used to select the final aliquot that is to be analyzed must be such as to meet the necessary resolution between the results based on the sampling mission objectives. Furthermore, the sampling parameters (depth and area) must be such as to give reproducible results. Michels (1971), in an analysis of data from around the Rocky Flats Plant concludes that Poet and Martell's (1972) sampling techniques probably introduced artificial variability in their results due to inadequate sampling depth. This made it difficult to differentiate between plant and world-wide contamination.

SOIL SAMPLING TECHNIQUES

Two basic techniques are presented in AEC (1974). The techniques are generally referenced to their developers; i.e., the HASL and NAEG techniques. They have similar philosophies, and generally are supplementary in that they are applicable for different soil types. The site selection criteria, outlined below, are similar for both techniques:

1. Select general sample locations based either on general areas around a site, average geographical distribution, or on a random basis (random numbers referenced to a geographical grid).
2. Pick undisturbed sites for actual sampling. This may require abandoning certain sites if the selection of general location is based on random numbers. Although usually unacceptable, disturbed sites, blow sand, dams, or recent landfill may be appropriate for certain mission objectives.
3. Pick open, generally flat areas where there are no nearby potential anomalies, such as near buildings or trees. Also avoid stream beds, dry wash bottoms, and hillsides.
4. Pick areas away from rock outcrops and with generally uniform vegetation coverage. Try to insure that the soil grain size is compatible with the sampling method.
5. Soil having high earthworm activity should be avoided due to the abnormally high vertical mixing.
6. Locations should be roughly 120 m (400 ft) or more from dusty roads or sites of previous construction.

The following items outline the HASL technique:

Surface Sample

1. Obtain surface samples by core technique. Any type of sampling tool that can remove an intact plug (cookie cutter-type instrument) is appropriate.
2. The surface sample depth should be 5 cm (2 inches). The sample area should be 500 to 1,000 cm^2 (about 0.5 to 1 ft^2). In grass areas the vegetation should be close-clipped and taken as a vegetation sample or discarded.

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* Energy Research and Development Agency, Health and Safety Laboratory, and Nevada Applied Ecology Group.

3. It is suggested that the sample be composed of about 10 plugs from a 5-meter line transect. The line should be located by reference to fixed landmarks.

4. The soil sampler should be pressed into the ground without twisting or disturbing the grass cover or soil surface.

5. The 5-cm depth is intended to include the soil of maximum plutonium activity and most of the root mat in areas covered with grass. In areas with a deeper root mat, it may be necessary to take a deeper surface sample to allow accurate estimation of the sample depth.

Depth Profile

1. A 3.5-in. diameter auger may be used to take incremental depth profile samples at the same locations where the surface plug was removed. Ten cores should be composited for a profile. The purpose of the profile determines the number of profiles that should be taken at a given location. Both the HASL and NAEG techniques recommend compositing a number of profiles (e.g., 10), but specific study objectives, such as determination of the movement of plutonium through the soil column, are best based on individual samples.

2. For the trench-type method, the vegetation, if present, is closely clipped, and the sod layer removed from the proposed trench area. A trench approximately 60 x 90 cm, and 60 cm deep is dug adjacent to the clipped area.

3. A rectangular three-sided 15x15x5-cm deep pan is used to take samples from the vertical wall of the trench.

4. A flat-bladed knife should be used to score the soil around the edges of the pan to allow removal of a sample having an accurate area.

5. The soil is removed from each side of the sampled area to provide a flat shelf prior to each 5 cm depth sampling increment.

6. The minimum profile depth should be based on analysis of preliminary samples (roughly a minimum of 20 cm).

7. The sampling area for this type of profile is only 230 cm^2, which provides a less representative deposition sample than does the surface sampling technique.

Sample Preparation

1. Spread out and air dry sample for about 3 days. Break up soil aggregates, and pull apart and cut up root mat. Weigh the total sample.
2. Remove and discard rocks greater than about 2.5 cm diameter. For gravelly soil, sieve through 10 mesh, removing material greater than 2 mm. Crush and blend sample.
3. Spread sample and quarter. Take a three-kilogram composite by taking small repetitive aliquots from each quarter and pulverize or grind this subsample.

The following items outline techniques patterned after those developed by the NAEG, but which were modified for the purposes of the Regulatory Guide 4.5 (AEC, 1974).

The NAEG techniques and sampling philosophy are described by Fowler, Gilbert, and Essington, "Sampling of Soils for Radioactivity: Philosophy, Experience, and Results; ERDA Symposium Series 38 (1974, CONF-740921.

The techniques were designed to be applicable to sandy soils, but more importantly were designed to minimize the potential of sample cross-contamination that can occur with coring techniques (smear of surface activity to subsurface sample). The NAEG does not generally composite samples, and neither endorses nor disapproves of transect sampling or compositing of samples. The specifications for compositing samples are based on the modifications of the NAEG techniques for the regulatory guide.*

Ring Method for Surface Samples

This technique can be used to collect either surface or profile samples.

1. A 12.7 cm-ID x 2.5 cm-deep ring is pressed into the soil. The soil inside the ring is removed with a disposable plastic spoon.
2. The soil from the outside of the ring is removed, and the ring is pressed down for another sample.
3. A surface sample is defined as a minimum depth of 5 cm. A minimum number of five separate samples should be taken along a straight line transect and composited for analysis.

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* Changes made subsequent to the original publishing of this report as ORP/LV-76-5 in 1976 (Reference Fowler, E. B. and E. H. Essington, Sampling Soils for Transuranic Nuclides: A Review: NAEG Symposium in Gatlinburg, TN, 1976, NVO-178).

Trench Technique for Surface or Profile Samples

1. Dig trench of convenient size, 15 to 25 cm deeper than desired sampling depth.
2. Take samples from trench wall with three-sided rectangular tray (10 x 10 x 2.5 cm deep).
3. Push the tray into the trench wall. Use a flat trowel to close the open end of the tray.
4. Remove the soil around the tray down to the sampling depth. Remove the sample.
5. A surface sample consists of soil taken from a minimum depth of 5 cm.
6. A minimum of five samples should be taken from separate trenches along a straight line transect. Composite the samples for analysis.

General Comments

1. Samples should either be double bagged or placed in cans.
2. Varying soil types require modification - Rocky soils may require larger samples to minimize the errors associated with sampling accurate areas and depths.
3. Locations should be identified by reference to fixed landmarks.
4. Adding moisture (as a fine spray) to the soil may minimize sampling problems.

Sample Preparation

1. Oven dry soil for 24 hr at 100°C. Weigh total sample.
2. Sieve sample to remove material greater than 0.6 cm. (0.25 in.) diameter (1/4-in sieve). This excludes rocks and most root material from further consideration.
3. Rocks can be acid washed, with the wash solution added to the solubilized soil sample.
4. Roots and vegetative material can be analyzed separately.
5. The sample should be ground (ball-milled) and blended prior to taking a representative aliquot for analysis.

Table 4 summarizes collection techniques used by several investigators.

The following points emphasize the similarities of the various techniques and potential pitfalls:

1. Both the HASL and NAEG techniques have standardized on a minimum depth for surface samples of 5 cm (2 in). For most locations and situations the majority of the plutonium is in the top 3 to 5 cm. A sample representing a depth of less than 5 cm may not account for the majority of the plutonium deposition (Krey and Hardy, 1970). Furthermore, the fractional uncertainty in the sampled depth is proportional to the sampled depth (e.g., a 1-cm uncertainty is 100% of 1 cm, but only 20% of 5 cm). An unnecessarily large depth results in diluting the higher surface concentrations with (usually) relatively uncontaminated soil. This increases the uncertainty in sample results. Mixing of surface soil with subsoil can also result in a significant scatter or variance in results, if uniform methods are not used.

2. AEC (1974) emphasized that when sampling rocky soils, modified techniques may be necessary. However, a representative depth is more important than a representative width. But as Bliss (1973) notes, the measurement of the cross-sectional area is more important than the measurement of the depth, because of the direct dependence of the deposition calculations on the area represented by the sample.

3. The AEC (1974)techniques emphasize the compositing of a number (five) of small-area samples for a given site to obtain a representative sample and a minimum sampled area (0.5-1 ft^2). This is not emphasized in many of the techniques in Table 4. In fact, as noted in Table 4, Bliss (1973) only composites two samples per horizon in depth profile samples. Bliss (verbal communication) indicated that REECO ususally only takes one sample per horizon. Moore, Office of Radiation Programs - Las Vegas Facility (ORP-LVF) (verbal communication) noted that probably only one sample per horizon was taken for Enewetak.

Several potential uncertainties are associated with sample preparation. These include:

1. Oven drying at 120°C versus air drying for several days. The differences in the resulting weights (up to roughly 10 percent) are present in the pCi/g values, but should be accounted for in the pCi/m^2 values.

TABLE 4. SAMPLE COLLECTION TECHNIQUES
(Blanks indicate no information)

		SURFACE SAMPLES								PROFILE SAMPLES							
Reference	Organization	Technique	Sample Area (cm^2)	Sample Area (in^2)	Number of Composites	Total Area (cm^2)	Total Area (in^2)	Sampled Depth (cm)	Sampled Depth (in)	Technique	Increments (cm)	Area Per Sample (cm^2)	Area Per Sample (in^2)	Number of Composites	Total Area (cm^2)	Total Area (in^2)	Sieve Size (cm)
AEC Guide (1974)	(HASL)	Core/Auger	60		10	600		5		Auger	0-30 total	62		10	600		<2.56
AEC Guide (1974)	(HASL)									Trench/Tray		225		1	225		<2.5
AEC Guide (1974)	(NAEG)	Ring	127		5	600		5		Trench/Tray	2.5-5	100		5	500		<0.2
AEC Guide (1974)	(NAEG)	Tray	100		5	500		5									<0.2
Bliss (1973)	NERC-LV/NAEG	Tray	100		10	1000		5		Trench/Tray	2.5-5	100		2	200		<0.2
Douglas (ORP-LVF)	ORP-LVF	Tray	100		10	1000		5		Trench/Tray	2.5-5	100		2	200		
Bliss (Verbal)	REECo/NAEG	Ring	127		≈1	127		5		Trench/Tray		≈100		≈1	≈100		
AEC (1973)	Enewetak	Core	30 or 60		1 or 2	30 or 60		5 or 15		Trench/Tray	≈5	100		1	100		
Corley, et al (1971)	Hanford	Shovel	549	216				1.3	0.5								
Corley, et al (1971)	Hanford	Tape Cont.	64		≈1	≤100		1.6									
Kahn, 10/1/74	EPA/NEF Cinn.							2.5	1								
Little (1973)	Colorado State University	Trench	25		4	100		3		Trench	3-21	25		4	100		
Poet & Martell (1974)		Spatula	1000		1	1000		1			0-0.3, 1.3, 2.5 ...	≈1000		1	≈1000		<.05
McClendon (1975)	SRP	Core	45		10	450		15		Core	0-5, 5-15, 15-22.5-30	45		10	450		
Krey & Hardy (1970)	HASL	Auger/Core	62		10	620		20		Auger/Core	0-5, 5-20	62		10	600		
Krey & Hardy (1970)	HASL	Template	930	144	1	930	144	20									
WASH-1259 (AEC, 1973)	LASL	Core	44		5	220		5									
WASH-1259 (AEC, 1973)	SRP	Core	62(?)		10	600(?)		5		Auger	0-5, 5-15, 15-22.5-30			10	600		
WASH-1259 (AEC, 1973)	Mound	Core	62		10	600		30.5	12								
WASH-1259 (AEC, 1973)	Mound	Scrape						0.3	1/8								
WASH-1259 (AEC, 1973)	Pantex	Core	62		≈1	≈62		5.1									
WASH-1259 (AEC, 1973)	Rocky Flats	Core	100		1	100		5									
WASH-1259 (AEC, 1973)	Sandia, Abq.	?	230	144	1	230	144	2.5	1								
WASH-1259 (AEC, 1973)	Argonne	Core	87		2	173		30									
WASH-1259 (AEC, 1973)	Idaho, NRTS	Core	79		5	390		5									
WASH-1259 (AEC, 1973)	ORNL	Core	46	7	9	410		1									
WASH-1259 (AEC, 1973)	Hanford							2.54	1								
WASH-1259 (AEC, 1973)	LLL							1;up to 25									

2. AEC (1974) refers to calculating field bulk density. This apparently is an error in semantics because the actual field weight is not obtained. Also, standard soil sampling methods are not used to determine the volume of hole from which the sample was taken.

POTENTIAL SAMPLING ERRORS

The available literature indicates that very few efforts have been made to evaluate the adequacy of soil sampling techniques. The following subjects will be treated in this section: consideration of apparent inconsistencies in results based on calculated bulk densities; consideration of plutonium depth profiles; discrete particles; and comparision of actual analytical variations in several groups of results.

Bulk Density

Kaufmann (internal memorandum dated October 3, 1974, ORP-LVF)*, noted that the tray/trench method was probably not adequate for obtaining an accurate estimate of sample weight per sampled area. This corresponds with a preliminary workup of data by Douglas (ORP-LVF, unpublished data) for the 1973 Trinity Field Study. Figure 2 shows a histogram of weight per unit area sampled and calculated bulk densities (g/cm^2). Histograms of other samples from this study indicate similar distributions. The maximum observed values from the Trinity study are equal to the minimum bulk density indicated by Terzaghi and Peck (1968) for uniform loose sand (1.43 g/cm^3). The median values indicated by Douglas for Trinity are roughly 30 percent lower than the value indicated by Terzaghi and Peck. Bliss (verbal communication) also indicated that values of less than 1 g/cm^3 have been noted in the Nevada Test Site (NTS) EMSL work.

Kaufmann (verbal communication) indicated that although valid values of about 1 g/cm^3 are not impossible, they are improbable. In nature they result largely from undisturbed drying of a saturated soil, forming an unconsolidated matrix-like material.

The American Society of Testing Materials soil sampling method D-1556 (ASTM, 1964) specifies a minimum sample size of 1400 cm^3 for soils with a maximum particle size of 12.5 mm or 0.5 inch diameter. Furthermore, the standard specifies a technique for measuring the sample volume by refilling the sampled hole with a measured weight of sand of known density.

The methods of Bliss (1973) and Douglas, which basically follow the NAEG technique, only collect about 1,000 cm^3 per horizon for profile samples--actually only 250-500 cm^3 per sampling cut. Furthermore, the use of the tray disturbs the actual sample and the surrounding area. Also, Bliss (verbal communication) notes that the two samples for profiles (NTS) are usually taken immediately adjacent to each other. Thus, the soil disturbed by the first cut is sampled in the next cut. Also, one side of the tray is not confined by soil during the second cut. There would appear to be inaccuracies associated with this type of sampling methodology. The significance of these is hard to assess, but could amount to 30 percent or more.

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*ORP-LVF, Office of Radiation Programs, Las Vegas Facility

Inaccurate bulk densities do not necessarily affect the results. The actual calculation is activity per unit weight times weight collected, divided by area sampled. The pertinent question relates to the representativeness of the grams of sample to the sampled area. Minor variations in the sampled depth probably have more affect on the interpretation of the results than on the actual numerical values.

Terzaghi and Peck (1968) present information on compressibility and the hysteresis loop after removal of the compression force for soil. A force of about 10-20 pounds applied to a scoop with frontal area of 50 cm^2 (10 cm x 5 cm) may produce a change in the void ratio (e) of up to about 10 percent of its value. The void ratio is the ratio of the volume of voids to the volume of soil substance. By relating the change in e to the change in porosity, n, ($n = e/1+e$), the change in the field bulk density of the soil can be estimated.

If the cutting edges of the scoop are assumed to transmit the force as a compression force to the total frontal area of the scoop, the bulk density at the frontal interface of the scoop is increased by roughly 5 percent. But part of the compressed soil would be in the scoop, and the compression would be reduced with distance from the scoop frontal interface. Thus, the maximum reasonable error would be less than 5 percent. This error would appear as a reduction in the actual amount of soil taken as a sample.

The bulking of the soil, as it is disturbed by inserting the scoop, tends to make it mound up in the scoop. Unless this is recognized, the tendency would be to only take a deep enough sample to fill the scoop, thus underestimating the volume sampled by about 20 percent. Bulking can also cause losses of material while taking the sample to be overlooked. Data from Terzaghi and Peck (1968) indicate potential errors of up to about 20 percent.

The EMSL-LV program has incorporated the use of scoops having an extra 2 cm length (10 cm sampling length, plus 2 cm for bulking, etc.) to minimize bulking and compression errors.

Taking profile cuts adjacent to each other could result in errors of roughly 10 percent, due to the disturbed nature of the soil and thus reduced bulk density, even if extreme care is taken in positioning the scoop on the open face of the second cut.

All of these errors tend to minimize the amount of sample actually obtained from an assumed area sample. This is apparently illustrated by the histograms of apparent soil density for the Trinity results as shown in Figure 2.

An experiment was conducted by EPA - Las Vegas staff to obtain an indication of variations in the soil volume collected by the scoop technique for depth profiles. Samples were taken by three experienced teams from a 10-meter diameter circle of relatively undisturbed desert. Two of the teams (A and B) basically used the NAEG scoop profile technique. The third team (C) used a displacement technique where the volume of soil removed was measured by filling the hole with a known volume of a standard density sand.

Team A actually took two side-by-side scoops (10 x 10 x 5 cm deep) from a trench. Not only were the scoops taken side by side, but a bench was not cleared off before going to the next lower depth. Team B took a single scoop. A bench was not cleaned off before sampling at the next depth.

Each team took four profiles. The only significant error noted was the sampling depth. Team A sampled to a depth of 22.9 cm versus the design depth of 20 cm.

Team B sampled to depths of 21.6 to 22.23 cm. These depth errors are equivalent to bulk density errors of about 10 percent. However, assuming the errors were generally uniform and that the actual sampling depths were measured, the bulk densities can be corrected.

The average bulk densities (wet weight) for the four profiles for teams A and B were 1.70 and 1.62 g/cm^3. Correcting for the sampling depth gives values of 1.49 and 1.48 respectively. These values compare well to the value of 1.53 g/cm^3 for team C. The standard deviation for all three sets of data was about 0.05 g/cm^3, indicating overlap of the data.

An interesting speculation is that concern for bulking of the sample and fear of not taking an adequate depth appears to result in over-compensation. The sampling depth may be deeper than expected.

Significance of Sampling Depth

The sample depth increment has a significant impact on sample results, and is inherently related to the objectives of the sampling program. This is just as true for results reported as activity per unit soil mass as for those reported as activity per unit area. Plutonium is deposited on the surface of the soil. Through mechanical action, as well as water and earthworm movement, etc., it is mixed through the upper soil layers, down to 20 cm, or more. The relative concentration with depth varies

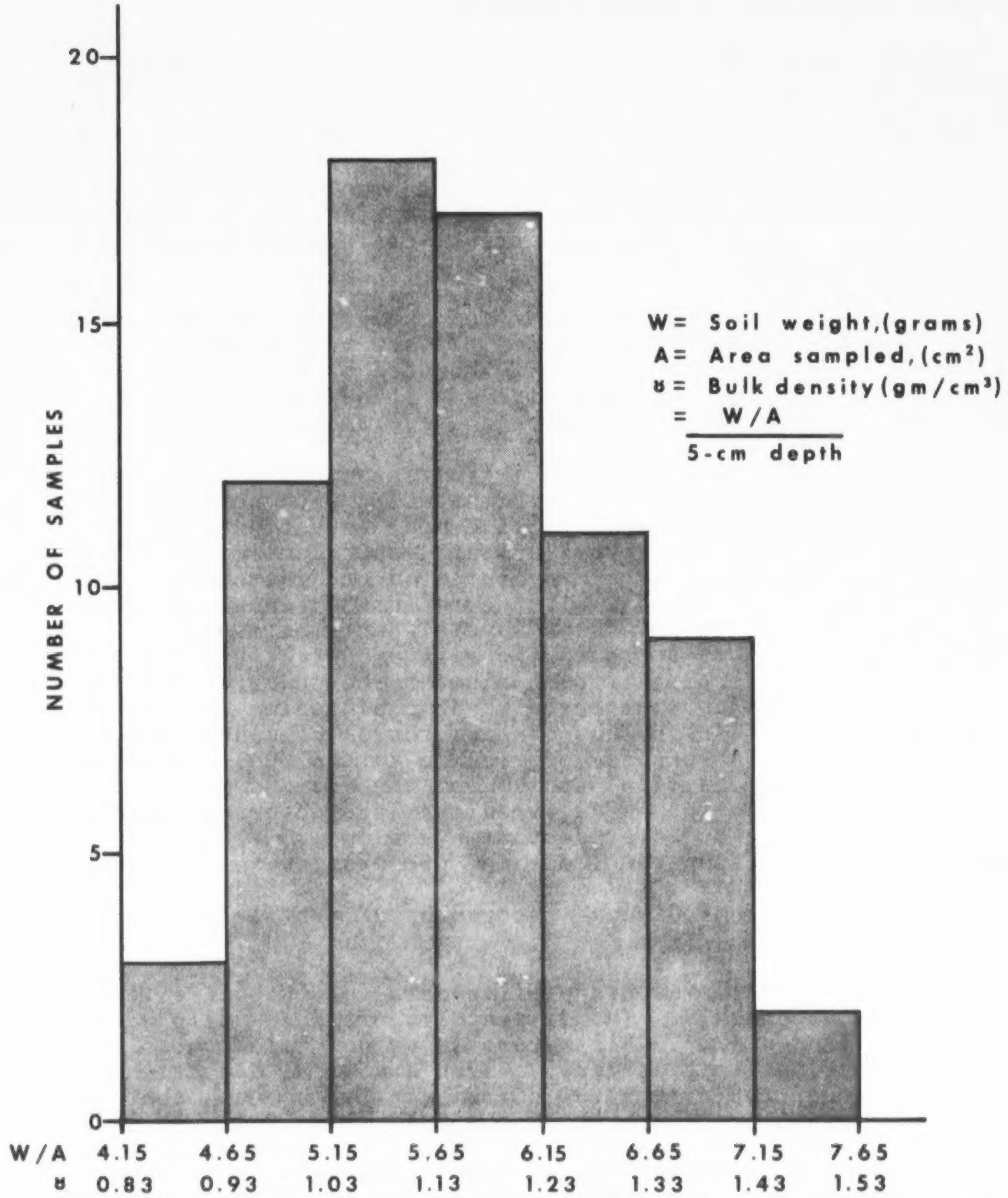

Figure 2. Histogram of weight per unit area for 72 soil samples from vicinity of Trinity, New Mexico. (from Douglas, EPA/ORP-LVF, unpublished data)

within localities and from one geographical area to another. Leopold et al. (1966), Colby (1963), EPA (1973a), Chepil (1945a, b,c,d), Chepil (1946), and Chepil and Woodruff (1963) present information on soil denuding, transport, and erosion as a result of natural forces and of human land-use.

Data on plutonium soil profiles from numerous areas are summarized in Table 5. These data include results from Savannah River, Georgia; NTS, Nevada; Rocky Flats, Colorado; New York, New York; and Trinity, New Mexico. The range of results, means, and standard deviations are given for the various sites. This presentation inherently assumes the data are normally distributed. This hypothesis has not been tested. Given the range and scatter of the data, the summarization and treatment is presented only as a trend or indication.

It is readily apparent that some of the groups of data, such as those reported by Bliss (1973) for NTS are not normally distributed. The data are inherently bounded by a value of 100 percent, and a value of the mean plus one standard deviation exceeds the 100 percent accumulation in several instances.

It is difficult to obtain meaningful information from some of the data because they are reported as pCi/g with no indication of the bulk density of the soil. Since most investigations vary the vertical increment with profile depth, each sample represents an average of a composite over a different depth increment. The variation of soil bulk density with depth further complicates the comparison. Also, at depths below several centimeters, the plutonium concentrations approach the minimum detectable activity (MDA)*. Given the detection of plutonium at the lower depths in many profiles, it is apparent there is some plutonium down to about 20 cm in most cases. Thus, the plutonium concentration postulated for the MDA results (e.g., zero to the MDA) influences estimates of the percent of plutonium for the various soil strata. There is also the speculation that the observed concentrations of plutonium at lower depths may be due to cross-contamination during sample collection, preparation, or analysis.

The following items discuss the groups of profile data in Table 5.

1. Bliss (1973) presents profile data for the off-site area around NTS. The data are reported in pCi/g of dry soil and nCi/m^2. Bliss reports values below the detection limit as zero. Given a nominal detection limit of 20 fCi/g (0.02 pCi/g), it can be seen that the exclusion of values below the detection limit can have a significant impact on the cumulative percent deposition for locations having deposition near background--roughly 1 nCi/m^2. For example, for the Furnace Creek-1 sample, the only detectable result in the profile was

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*MDA (minimum detectable activity)

TABLE 5. PERCENTAGE PLUTONIUM DISTRIBUTION IN SOIL AS A FUNCTION OF DEPTH

Location	Reference	0-1 cm Depth				0-2.5 cm Depth				0-3 cm Depth				0-5 cm Depth				0-15 cm Depth			
		n*	$\bar{x}$*	S*	Range	n	$\bar{x}$	S	Range	n	$\bar{x}$	S	Range	n	$\bar{x}$	S	Range	n	$\bar{x}$	S	Range
Nevada Test Site, NV^2	Bliss (1973)	39	52	32	0-100%	-------				41	91	20	16-100%	-------				41	99	5	76-100%
Nevada Test Site, NV^3	Gilbert & Eberhardt (1974)	-------				-------				-------				13	97	3	90- 99%	-------			
Rocky Flats, CO	Krey & Hardy (1970)	-------				-------				-------				7	62	17	39- 91%	-------			
Rocky Flats, CO^1	Poet & Martell (1972)	7	52	38	8- 94%	7	69	33	17=100%	-------				5	83	27	39-~100%	-------			
Trinity Site, NM	Douglas	-------				-------				-------				4	50	24	27- 84%	4	79	22	46- 95%
New York City, NY	Krey & Hardy (1970)	-------				1	66	--	--	-------				1	81	--	--	-------			
Waynesville, OH	Krey & Hardy (1970)	-------				-------				-------				1	47	--	--	-------			
North Eastham, MA	AEC (1974$_a$) Wash-1359	-------				1	~53	--	--	1	~69	(@ 4 cm)		1	~77	(@ 6 cm)		1	~90	--	--
Savannah River, GA^2	AEC (1973$_a$) Wash-1259	-------				-------				-------				7	61	7	48- 73%	7^2	91	4	--

1. Depth intervals are missing from several profiles. The interval was 0-1.3 cm versus 0-1 cm. Deposition was calculated from the original data by assuming a bulk density.
2. Most of the profiles indicated undetectable plutonium levels below 15 cm.
3. Excludes two values of 38 and 46.6%. These values give $\bar{x}$ = 90; S = 20.

*n = number of samples

*$\bar{x}$ = mean Pu in increment, as percent of total deposition

*S = standard deviation

0.02 pCi/g. This then indicates 100 percent of the activity was in the first centimeter of soil.

If it is postulated that all the samples contain 0.01 pCi/g (about one-half the nominal MDA), the following profile is noted:

Depth (cm)	Bliss (pCi/g)	Postulated pCi/g	Calculated nCi/m *	Postulated Cum %	Bliss Cum %
0-1	0.02	0.02	0.3	9	100
1-3	0	0.01	0.2	15	-
3-7	0	0.01	0.5	29	-
7-15	0	0.01	1.2	65	-
15-23	0	0.01	1.2	100	-

* Calculated by assuming soil density of 1 g/cm^3 from 0-5 cm and 1.5 g/cm^3 from 5-23 cm.

The postulated values differ from Bliss' estimates, for this extreme example, by over a factor of 10. If a level equal to the MDA were postulated, the difference would be a factor of 20. The assumption of zero for MDA values can easily account for variances of tens of percent in the cumulative deposition. With a lower MDA, this effect would decrease.

The following items summarize specifics from Bliss (1973):

1. A large fraction of the total plutonium is generally in the top centimeter of soil.
2. The top 5 cm of soil generally contains over 90 percent of the detected activity.
3. Excluding one sample (Moapa-1), whose values are at or near the MDA, 50 percent or more of the detected activity is in the top 3 cm of soil.

Gilbert and Eberhardt (1974) summarize profile data from Areas 5 and 13 on the NTS. The following observations can be drawn from their data:

1. Thirteen of 15 profiles indicated over 90 percent of the detected plutonium was in the top 5 cm for desert

pavement areas. The other two areas gave values of 38 and 47 percent.

2. The average of the 13 values is given in Table 5. The mean for all the values is 90 percent with a standard deviation of 20 percent.

3. The averages for Areas 13 and 5 are similar. However, if the two low values are included, the mean for Area 13 is lower than that for Area 5. The two low values also cause a large increase in the standard deviation.

4. The authors conclude that most of the profiles have greater than 95 percent of the plutonium in the top 5 cm. The actual data are not presented, so the presence and treatment of MDA values cannot be assessed.

5. The authors noted a trend toward a decrease in the plutonium:americium ratio with depth.

Krey and Hardy (1970) present profile data for Rocky Flats, New York City, and Waynesville, Ohio. The following points are noted:

1. Only about 62 percent of the plutonium was found in the top 5 cm.

2. As much as 60 percent was found below 5 cm.

Poet and Martell (1972) report data for the Rocky Flats area. Their profiles generally extended to only 10 cm or less; and several increments are missing in the reported data. Furthermore, the data were only reported in units of dpm/g. The data were transformed to units of nCi/m^2 by multiplying by the incremental depth of the sample and a postulated bulk density. The density from 0-5 cm was assumed to be 1 g/cm^3 (based on Poet and Martell, 1972, and random estimates derived from Krey and Harty, 1970). A density of 1.5 g/cm^3 was used for samples below 5 cm (estimated from Krey and Hardy, 1970).

Table 6 (data from Poet and Martell, 1972) indicates general uncertainties in the data as a result of the sampling techniques for the profiles (fractions of a centimeter to a centimeter), the previously mentioned transformation assumptions, point-to-point variations, etc.

TABLE 6. COMPARISON OF SURFACE AND PROFILE SAMPLES

Location	Profile Depth (cm)	Estimated cumulative deposition based on depth profile (nCi/m²)	Estimated deposition based on 1 cm-deep surface sample* (nCi/m²)
J	0-0.3	6.27	
	0-1.3	14.06	5.18
K	0-0.3	6.78	
	0-1.3	4.01	3.97
I	0-0.3	0.12	
	0-0.7	0.21	7.75 (Taken prior to profile)
	0-0.7 plus 1.3-2.5	0.45	

* Different data from profile sample

The variations between surface samples and profile samples from similar depths range from over two to greater than an order of magnitude. Poet and Martell (1972) note a large build-up of soil from wind erosion at Site I subsequent to taking these samples, which probably explains the apparent discrepancy for that site. However, it should be emphasized that this was not noted when the samples were first taken. This indicates the problems in taking characteristic samples--hindsight helps.

The following items characterize the Poet and Martell data:

1. About 52 percent of the detected plutonium was found in the first 1.3 cm of soil.
2. About 69 percent was found in the top 2.5 cm of soil.
3. About 83 percent was found in the top 5 cm of material. Given the calculated standard deviations, the range and limitations of the data, and the assumptions necessary to transform the data, the value of 83 ± 23% for Poet and Martell is considered similar (not statistically different even at low probabilities) to the value of 62± 17% for Krey and Hardy.

Data for the Trinity, New Mexico site are based on four samples taken in November 1973. The data will be published in a future ORP-LVF report. The concentrations ranged up to 1 pCi/g and 47 nCi/m^2.

Since actual MDA values were reported, the concentration was postulated to be equal to the MDA. The percentage of total deposition for 0-15 cm becomes 78 percent with a standard error of 16 percent. These values are essentially indistinguishable statistically.

Data from the Savannah River plant were transformed from pCi/g to nCi/m^2 as indicated previously. All but one of the profiles indicated values below the MDA for strata below 15 cm. Thus, the first value below the detectable limit was set equal to the MDA.

The profile from North Eastham, Mass. (AEC, 1974b) is from a background location.

The most general conclusion that can be drawn from the summary of profile data in Table 5 is that the initial phase of a soil sampling program should include profile samples to characterize the area. Further, 5 cm is a prudent minimum depth for surface samples.

A non-weighted average of the values in Table 5 indicates that 72% ± 18% (1 sigma) of the activity is above 5 cm. Given the potential for bias in the various groups of data, a non-weighted mean appears to be reasonable. If the values in the table are weighted by the number of results represented by each value, the average is 83 percent.

Table 7, taken in part from Krey (1974) compares some of his data with that from Poet and Martell (1972). Krey notes that Poet and Martell's data are generally low by a factor of 10. This evaluation is based on Poet and Martell's data for the top 1 cm of soil, and Krey's data for a 20 cm sampling depth.

It should be noted that these data are very difficult to compare due to the difference in sampling depth, and possibly more importantly, Poet and Martell only report their data in pCi/g. If a surface soil density of 1 g/cm^3 (suggested by Poet and Martell, and used by Krey for the comparison) is used for the first 5 cm, and 1.5 g/cm^3 for the 5 to 15 cm increment, some of the Poet and Martell data can be related to the same general depth used by Krey (1974). Three values are presented in Table 7.

Poet and Martell (1972 and 1974) note that their objective was to detect the recent deposition of plutonium and indicate an inhalation hazard - thus their choice of a shallow sampling depth. In any case, although the original data in Table 7 indicate a significant disagreement between the two sets of data, the data for similar sample depths are generally compatible.

TABLE 7. COMPARISON OF PLUTONIUM SOIL SAMPLING DATA

Site	Poet and	Martell	Data	Krey & Hardy Estimate
	(nCi/m²) 1 cm surface sample	(nCi/m²) profile data	Profile depth (cm)	(nCi/m²)
A	5.8			15
B	10			35
	61			35
C	0.41			11
	0.26			11
I	7.7	3	14	4
J	5.4	15	10	20
K	4.0	11	2.5	14
L	0.52			17
M	1.7			17
N	6.0			30
V	1.4			4
V	2.4			8
W	0.15			17
Z	0.84			4

It appears, based on the comparisons in Table 7 of the Poet and Martell and the Krey data, and the similar tabulation in Table 6, that a 1-cm sample depth results in a large variation of the data. This is reflected by the large standard deviation noted in Table 5.

Discrete Particulate Material

Various authors (Poet and Martell, 1972, and Sill, 1971) have related variations between samples to discrete particles, whereas other authors relate variations to inadequate sample collection and aliquoting techniques (Krey and Hardy, 1970 and 1974). Sill (1971) and Sill and Hindman (1974) emphasize the limitation of various analytical techniques for complete dissolution of refractory plutonium particles. They not only indicate concern with insoluble refractory material in the original sample, but also with the formation of refractory material during sample preparation. This section is only concerned with the sampling implications of discrete particles. The analytical implications will be dealt with in another section.

Plutonium contamination in the environment does not appear to be in the form of discrete particles composed of plutonium oxides. Rather, soil and air particles containing plutonium appear to be composed of natural particles with plutonium oxides generally dispersed in the particle or natural particles agglomerated with one or more plutonium oxide particles (Nathans and Holland, 1971, and Bretthauer et al., 1974). The actual characteristics of the particles is expected to vary depending on the source of formation and release (e.g., plutonium in oil leaking from drum at Rocky Flats, and explosive detonations at NTS).

Table 8 indicates characteristics for various size particles.

TABLE 8. PLUTONIUM PARTICLE CHARACTERISTICS

Isotope	Diameter (μm)	pCi per particle	particle per pCi	Particle per gram of soil for 0.1 pCi/g	Particle per 930 cm^2 @ 1 nCi/m^2
^{239}Pu	0.1	0.000325	3076	308	286,000
	1	0.325	3.077	0.308	286
	1.5	1.096	0.912	0.091	84.8
	2	2.60	0.385	0.039	35.8
	5	40.59	0.0246	0.0025	2.29
^{238}Pu	0.1	0.09	11.1	1.1	1021
	1	90.99	0.0110	0.0011	10.2
	1.5	307	0.0033	0.00033	0.30
	2	728	0.0014	0.00014	0.13
	5	11,370	8.8E-5	8.8E-6	0.008

Ettinger et al. (1967), Mishima and Schwendiman (1970), Kelkar and Joshi (1970), Molen and White (1967), Sherwood and Stevens (1965), Hunt (1971), Mishima (1964), and Kirchner (1966) present data on particle size distributions expected and observed around various types of plutonium operations and accidents. The mean sizes vary from less than one to tens of micrometers (Mishima, 1964, and Mishima and Schwendiman, 1970). The most probable geometric mean sizes for release appear to be around 1 micrometer, with geometric standard deviations of about 1.5 to 3. Nuclear explosions apparently produce particles a few millimicrometers in diameter (Klement, 1965).

Although the agglomeration of plutonium particles to soil and dust particles changes the basic size distribution, the activity per aggregate particle should relate to the original plutonium particle or particles. Thus, for samples near facilities associated with plutonium releases, it is possible that the contamination is composed of two or more particle size distributions (worldwide fallout and facility) with one of the distributions in the micrometer size range. Thus, as noted by Sill (1971), the deposition near such facilities may be heterogeneous, when viewed from one-, 10-, or even 100-g samples. Table 8 shows the number of particles in a sample of given size.

If samples are based on a significant fraction of a 1000 cm^2 area (929 cm^2 per ft^2) the homogeneity of plutonium deposition within the area is less critical than is the homogeneity within the sample aliquot taken for actual analysis.

Table 9 presents a set of results from Sill (1971). Geometric means, $\bar{X}$, and geometric standard deviations, S, have been calculated for the various groups of data. The column on the right indicates the ratios of the maximum to minimum values for the 95 percent confidence range. The first two samples were collected from an area that should have only been exposed to global fallout. The values for the duplicate analyses reflect expected analytical variations (Sill, 1971). It should be noted that the background concentrations varied by a factor of two. This could possibly relate to different sample depths, in which case the deposition numbers (nCi/m^2) might have less variance.

The third through seventh samples (Table 9) were collected downwind of a facility where there was a known release. This is evident in the observed plutonium concentrations and general increase in the ratio of the 95 percent confidence limits, which indicates more heterogeneous distribution. Sample 7, which was 43 miles downwind, is an exception to both the concentration and heterogeneous distribution comments. Sample 4, although elevated, also indicates a fairly uniform distribution. Samples 8 and 9 are from another facility with a known release. The larger range for Sample 8 is due to only one result. The general scatter in the duplicate results for Sample 9 and the range in the 95 percent limits reflect that it was collected near the facility, apparently in an area of heterogeneous deposition.

Many of the variations in Table 9 can be accounted for by micrometer-size particles of PuO_2. The activity of plutonium particles is proportional to the diameter cubed of the particle. Thus, using data from Table 8, the high result for Sample 3 (Table 9) could have been due to about one 1-micrometer particle

TABLE 9. REPRODUCIBILITY OF ANALYSES USING 10-GRAM ALIQUOTS OF PREPARED SOILS (From Sill, 1971)

Number	Measured Pu-239 (dpm/g)	Location or Type Sample	ln $\bar{x}^a$	ln S	t (95%)	$\bar{x}$	Ratio of Upper and Lower 95% Confidence Limits
1	0.110 ± 0.009 0.116 ± 0.010 0.112 ± 0.012 0.101 ± 0.008 0.111 ± 0.008	General Bkgd.	-2.21	0.051	2.776	0.11	1.3
2	0.060 ± 0.007 0.050 ± 0.007 0.054 ± 0.008 0.063 ± 0.007	General Bkgd.	-2.87	0.104	3.182	0.056	1.9
3	1.59 ± 0.04 0.56 ± 0.02 0.94 ± 0.03 0.68 ± 0.03	2 miles downwind of facility with Pu release.	-0.141	0.456	3.182	0.87	18
4	0.62 ± 0.02 0.56 ± 0.02 0.57 ± 0.02	2 miles downwind of facility with Pu release.	-0.540	0.054	4.303	0.58	1.6
5	0.044 ± 0.006 0.077 ± 0.008 0.042 ± 0.005 0.055 ± 0.010 0.047 ± 0.006	16 miles downwind of facility with Pu release.	-2.96	0.245	2.776	0.052	3.9

TABLE 9. REPRODUCIBILITY OF ANALYSES USING 10-GRAM ALIQUOTS OF PREPARED SOILS
(From Sill, 1971)
(Continued)

Number	Measured Pu-239 (dpm/g)	Location or Type Sample	$\ln \bar{x}$[a]	ln S	t (95%)	$\bar{x}$	Ratio of Upper and Lower 95% Confidence Limits
6	0.079 ± 0.009 0.058 ± 0.008 0.071 ± 0.009 0.29 ± 0.01	17 miles downwind of facility with Pu release.	-2.32	0.731	3.182	0.099	104
7	0.051 ± 0.007 0.066 ± 0.009 0.056 ± 0.006 0.052 ± 0.006	43 miles downwind of facility with Pu release.	-2.88	0.117	3.182	0.056	2.1
8	0.071 ± 0.008 0.22 ± 0.02 0.051 ± 0.007 0.059 ± 0.006	50 miles downwind of facility with Pu release.	-2.49	0.665	3.182	0.083	69
9	0.35 ± 0.02 0.78 ± 0.04 1.73 ± 0.04 0.26 ± 0.01	100 yds. downwind of facility with Pu release.	-0.524	0.852	3.182	0.592	227

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[a] The analytical error estimates have not been considered in the statistical summarization of the data.

per gram, or a single particle having a diameter of about 2.5 micrometers in the total 10-gram sample, above the global background.

As part of the NAEG program, a set of 20 soil samples were collected from Penoyer Valley, Nevada, about 20 miles northeast of the NTS. The samples were split into duplicates and two aliquots were taken from each duplicate. The scatter of results from these 80 samples was such that the variations between the four positions from an individual site and the 80 samples could not be related to a rational explanation of sampling or analytical errors.

In an attempt to resolve this problem, portions of the sample from one site were split for inter-laboratory analyses. Table 10 shows replicate aliquot analyses of the sample by three laboratories. Each laboratory used its standard analytical method to analyze aliquots of less than 10-mesh desert soil. Most of these data were published by Bliss (1973). Although the sample preparation and analysis techniques vary somewhat between the three labs, they are basically the same. The analyses were all done by the basic acid dissolution technique (HCL, HF, and HNO). The specific techniques vary, in part, because of the different sample sizes.

Although the geometric means from the different labs vary, all of the 95 percent confidence levels (C.L.) have a significant overlap. These data illustrate the dramatic decrease in the ratio of the extremes of the 95 percent confidence range with the increase in sample size.

The geometric mean and standard deviation for the 80 replicate results, and the 95 percent C.L. estimates and their ratios are given in Table 11. The mean and C.L. estimates and their ratios include the values from Table 10. This would be expected, since the group of 80 replicates is based on a sample from four sites, whereas the interlaboratory samples came from one of the four sites. A frequency distribution table and probability plot for the 80 values is shown in Appendix C.

Means, confidence levels, and ratios of the C.L. are given for three other groups of data in Table 11 (Bliss, 1973). The samples from Baker, California and Kingman, Arizona and northwest of NTS indicate background values and have much lower C.L. ratios than the other two samples. The Baker and Kingman group of data represents several sites, and thus would be expected to have a larger range than the data from the location northwest of NTS.

TABLE 10. COMPARATIVE ANALYSES OF PLUTONIUM-239 IN SOIL

Lab	Aliquot Size (g)	Pu-239 (pCi/gm)	ln $\overline{x}$	ln S	$\overline{x}$	Range 95% C.L.	Ratio of Upper and Lower 95% Confidence Limits
EPA	1	0.23	0.405	0.952	0.67	0.085-5.21	61
	1	0.24					
	1	0.27		(n = 14; t = 2.16)			
	1	0.37					
	1	0.30					
	1	0.40					
	1	0.42					
	1	0.53					
	1	0.67					
	1	1.0					
	1	1.2					
	1	1.4					
	1	3.0					
	1	5.3					
REECo	10	0.66	0.646	0.674	1.9	0.415-8.76	21
	10	0.90					
	10	1.3		(n = 10; t = 2.26)			
	10	1.4					
	10	1.5					
	10	2.0					
	10	2.6					
	10	3.3					
	10	4.4					
	10	5.2					

TABLE 10. COMPARATIVE ANALYSIS OF PLUTONIUM-239 IN SOIL (Continued)

Lab	Aliquot Size (g)	Pu-239 (pCi/gm)	ln $\bar{x}$	ln S	$\bar{x}$	Range 95% C.L.	Ratio of Upper and Lower 95% Confidence Limits
LLL	25	1.9	1.078	0.6312	2.9	0.97-10.32	11
	25	4.6					
	25[a]	3.3	1.226	0.0415	3.4		
	25[a]	3.5					
	100	4.1	1.565	0.213	4.8	2.33-10.4	4.5
	100	5.5					
	100[a]	4.0	1.635	0.3374	5.1		
	100[a]	6.5					

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[a] Aliquot received additional grinding and blending prior to analysis.

TABLE 11. PERTINENT STATISTICS FOR Pu-239 RESULTS FROM SELECTED SAMPLE GROUPS (from Bliss, 1973)

	Location and Units of Activity			
	Baker, CA and Kingman, AZ (nCi/m^2)	Penoyer Valley Replicate Site (pCi/g)	Northeast of NTS (nCi/m^2)	Northwest of NTS (nCi/m^2)
No. of Results	27	80	100	35
ln X	0.068	0.053	1.881	0.584
ln S	0.869	1.336	1.261	0.572
X	1.071	1.054	6.557	1.793
Lower 95% C.L.	0.188	0.0729	0.527	0.571
Upper 95% C.L.	6.086	15.245	81.638	5.627
Ratio:				
Upper to Lower C.L.	32	209	155	10

The sample from northeast of NTS is at the extreme of or above background. The Penoyer Valley results are generally indicative of roughly 10 nCi/m^2 or higher. Thus, both of these samples appear to contain dispersed global fallout, plus rather discrete NTS fallout.

The variation in the analyses of these samples is relatable to a variance of one or several particles, of one to several micrometer diameter, per gram of sample. The actual numbers depend on the particle size and sample size of concern. The potential variation in results for 1-gram samples is particularly obvious. A single one-micrometer particle can cause a multiple variance in results. This would give strong credence for taking a minimum sample aliquot of 10 grams for analysis.

Little et al. (1973) present a limited amount of data for soil grain size in the Rocky Flats areas and the percent of plutonium associated with the various grain size increments. The plutonium concentration, pCi/g, for two samples is inversely proportional to the soil grain size, 0.1 to 5 mm. Tamura (1975) presents similar data (see the end of this section).

The heterogeneous deposition of plutonium-238 presents an even greater problem than for plutonium-239, because the specific activity of plutonium-238 is about 280 times that of plutonium-239. Furthermore, the concentration of plutonium-238 in the environment is normally much lower than that of plutonium-239. Plutonium-238 background levels are roughly 1 fCi/g for soil samples several cm deep (Krey and Hardy, 1970, and Robinson et al., 1975). Thus, from Table 8 it can be seen that a plutonium-238 dioxide particle of one micrometer diameter in a 10-gram sample can give a value of 9 pCi/g, or four orders of magnitude above background. Even a 0.1-micrometer particle in a 10-gram sample gives a value of 9 fCi/g.

Robinson et al. (1975) report results of two programs where samples were split between Mound Laboratory and EPA. The ratios of the results of these programs are shown in Table 12. The samples collected by EPA were split in the field. The samples taken by Mound Laboratory were first dried and ground to less than 20-mesh particle size. It is evident from Table 12 that the samples split after mixing gave more comparable results than did those which were split in the field.

TABLE 12. INTERLABORATORY COMPARISON OF MOUND LABORATORY AND EPA RESULTS OF PLUTONIUM-238 IN SOIL AND SEDIMENT (From Robinson et al., 1975)

Samples Split in Lab

Code	Mound (nCi/g)	EPA (nCi/g)	Ratio of Results Mound/EPA
EA1	0.0001 ± 0.0001[a]	0.00011[b]	0.91
EB1	<0.0001[a]	0.00012[b]	< 0.83
EC1	0.0029 0.0011	0.0048	0.60
ED1	0.0009 ± 0.0004	0.0011	0.82
EE1	0.425 ± 0.024[a]	0.440	0.97
EF1	1.03 ± 0.05[c]	1.13[b]	0.91
EG1	0.0098 ± 0.0027	0.0108	0.91
EH1	0.0238 ± 0.0053	0.026	0.92
EI1	<0.0001	0.00098	< 1.02
EJ1	0.0010 ± 0.0005	0.0011[b]	0.91
FA1	0.0094 ± 0.0026	0.0085[c]	1.11
FE1	0.0138 ± 0.0025[b]	0.0181	0.76
GA1	0.0004 ± 0.0002	0.00048	0.83
HA1	0.0047 ± 0.0016	0.0051	0.92
IA1	0.0020 ± 0.0008	0.0025	0.80
JA1	0.0007 ± 0.0004	0.0007	1.00
KA1	0.0309 ± 0.0065	0.027	1.14
LA1	0.0096 ± 0.0027	0.0109	0.88
CE1	0.0302 ± 0.0064	0.024	1.26
QE1	1.00 ± 0.09	0.920	1.09
			n=20 $\bar{x}$=0.929 S=0.147

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[a] Mean of quadruplicates
[b] Mean of duplicates
[c] Mean of triplicates

TABLE 12. INTERLABORATORY COMPARISON OF MOUND LABORATORY AND EPA RESULTS OF PLUTONIUM-238 IN SOIL AND SEDIMENT
(From Robinson et al., 1975)
(Continued)

Samples Split in Field

Code	Mound (nCi/g)	EPA (nCi/g)	Ratio of Results Mound/EPA
EPA-17	0.284 ± 0.035	0.047	6.04
EPA-18	0.280 ± 0.035	0.060	4.67
EPA-1	0.165 ± 0.023	0.230	0.72
EPA-6	0.0052 ± 0.0017	0.0038	1.37
EPA-20	0.0011 ± 0.0005	0.0019	0.58
EPA-14	0.0009 ± 0.0004	0.00044	2.05
EPA-15	0.0009 ± 0.0004	0.00096	0.94
EPA-3	<0.0001 ± 0.0001	0.00039	<0.26
EPA-13	<0.0001 ± 0.0001	0.00010	<1.00
EPA-7	<0.0001	0.00044	<0.23
EPA-12	<0.0001	0.00019	<0.53
EPA-2	<0.0001	0.00012	<0.83
			n=12 $\bar{x}$=1.60 S=1.84
ALGAE SAMPLES			
EPA-9	0.111	0.079	1.41
EPA-21	0.0024	0.00088	2.73
			n=2 $\bar{x}$=2.07 S=0.93

PARTICLE SIZE DISTRIBUTION OF PLUTONIUM IN SOIL

Although inhalation is generally considered to be the primary intake pathway for plutonium, soil is generally considered to be the primary reservoir of environmental contamination. Thus, there is a need to relate soil sample results to potential or actual airborne concentrations. The first part of this section has addressed techniques primarily intended to quantitate the amount of plutonium in soil. Thus, the emphasis has been to take samples of a reproducible and sufficient depth in order to assess the total plutonium inventory. Sampling for resuspendible plutonium requires different priorities and considerations. Presently applied techniques include a one-eighth inch depth sample by the State of Colorado, 1-cm depth samples by Poet and Martell (1972), and techniques using sticky paper placed in contact with the soil surface (Volchok, 1971).

More recently, McLendon et al. (1975) published results where a vacuum cleaner type instrument was used to collect the resuspendible material from the area of the sample head. This technique appears to have merit, but sample results have not been directly related to air concentrations.

Johnson et al. (in press) proposed that the less than 5-micrometer (density 11 g/cm^3; i.e., 17 micrometer density 1 g/cm^3)*size material that can be swept from the soil surface be used as an indication of inhalation hazard. The sample fractionation procedure includes breaking the soil down to basic discrete particles. Thus, the technique would appear to reverse the "weathering" effect that decreases the relative resuspendibility of old versus newly deposited contamination (Anspaugh et al., 1975).

There presently is no accepted technique for measuring resuspendible material from soil. However, data from several studies allude to soil being associated with various particle size fractions (Johnson et al., Little et al., 1973, and Tamura, 1975). Since resuspension is dependent on the soil particle size distribution (Anspaugh et al. 1975), as well as other factors, the size distribution of plutonium in soil is considered to be pertinent basic information.

The ORP-LVF obtained several samples from Rocky Flats to independently investigate the size distribution of plutonium in the soil. The samples were collected by the Rocky Flats Environmental Research and Development Administration area office several hundred yards downwind of the pad where the basic Rocky Flats plutonium contamination incident originated. Although it was originally presumed that the samples would contain less than about 25 pCi/g of plutonium-239, they actually contained over 500 pCi/g. Thus, because of concern for laboratory contamination, they received less extensive analysis than originally proposed.

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*The use of a density of 1 g/cm^3 is based on the definition of the equivalent aerodynamic diameter. Conversions to equivalent diameters in this section are based on the settling velocity in air--see next section.

A sample from about 35 miles downwind at the Trinity, New Mexico site was also analyzed.

The Rocky Flats samples were collected in February 1976 from the surface centimeter of soil, from an area of 2000 cm^2. The Trinity sample was collected in December 1974. It represented the surface 2.5 cm of soil from an area of 2500 cm^2.

Two Rocky Flats samples were partitioned into three separate aliquots. The aliquots were further partitioned as described below (the aliquots are denoted as A, B, and C in Table 13).

1. Dry sieve through 10-mesh sieve--Less than 2 mm
2. Wet sieve (not dried material) through 140-mesh sieve--Less than 102 micrometer
3. Elute material from sedimentation column for a Stoke's Law setting velocity of less than 3.37×10^{-3} cm/sec--(10 micrometer at 1 g/cm^3, or 6.1 micrometer at 2.65 g/cm^3; Krumbein and Pettijohn, 1938)

The samples were prepared according to the general techniques presented by Folk (1961). The following comments relate to specific information on the sample preparation procedure:

1. Radiochemistry was performed on about 10 gram aliquots of the samples.
2. The samples were not completely dried prior to sieving (forms and increases the stability of conglomerates, Falk, 1961).
3. A solution of Calgon, sodium metaphosphate, was used as a dispersing agent for the sedimentation separation (about 10 ml of a solution of 40 g/liter).
4. The material retained on the 10-mesh sieve was washed with a solution of Calgon, and the wash included with the less than 10-mesh material.
5. The material retained on the 140-mesh sieve during wet sieving was then dry-sieved through the 140-mesh sieve. Folk (1961) notes that the fines are partially bound to the coarse material by moisture bonds during wet sieving. The amount of material passing 140 mesh was increased by 4 to 10 percent for the Rocky Flats samples and 20 percent for the Trinity sample. This material was not used in the mass balances or for radiochemistry analysis.

6. The "pipet aliquoting" procedure for determining the total fraction of the sample less than 10 micrometer was incorrectly done on oven-dried samples. Thus, the results were anomolously low. The values for the total sample fraction less than 10 micrometer are therefore estimates based on multiple elutions from the settling column. Various numbers of elutions indicated values of up to 10 percent for the Rocky Flats samples. Based on the material recovered, the total value is estimated to be 20 percent.

7. The amount of the plutonium in the greater than 2-mm size fraction was not determined.

The results of the study are given in Table 13. The size fractions of less than 2 mm and 100 micrometers are based on the sample that passed 10- and 140-mesh sieves, respectively. The less than 10 micrometer size is passed on an equivalent aerodynamic diameter in air (density of 1 g/cm^3).

The Rocky Flats soil had a smaller particle size distribution than the Trinity sample. The size difference is also reflected in the distribution of plutonium. About 50 percent of the plutonium from the Rocky Flats samples was associated with the less than 10-micrometer size versus about 10 percent for the Trinity sample. The specific activity of plutonium in soil (pCi/g) appears to be generally inversely proportional to particle size. The ratio of the concentration of plutonium in the less than 10-micrometer fraction to that in the less than 2-mm fraction (basic soil size) was about 2.4 versus 1.8 for the less than 100-micrometer size fraction.

The radiochemistry results indicate good reproducibility for the preparation and analysis procedures. The only anomalous result appears to be the value of 1580 pCi/g for 1A (less than 100 micrometer). The other results for the various size fractions are within the two-sigma counting errors.

The mass fractions also show reasonable reproducibility for the sample preparation procedures. The mass of the less than 10-micrometer size fraction varies because a varying number of sedimentation runs were done for each sample. The fraction of material in the less than 10-micrometer size range is based on the maximum amount of material recovered (sample 2A) and a subjective observation that about half of the available material was recovered.

The results are compared to those of other investigators in Table 14. In general, the Trinity results for mass fractions are similar to those of Tamura (1975) for NTS (similar sandy soils). The results of Johnson et al. show reasonable agreement with the ORP-LVF results, especially considering the differences in the treatment techniques. Johnson et al. used hydrogen peroxide to

TABLE 13. SOIL MASS AND PLUTONIUM ASSOCIATED WITH VARIOUS PARTICLE SIZE FRACTIONS OF SOIL

	Mass and Activity of Material				Fraction of Material Passing 10 Mesh Sieve		
Sample and Units	Greater Than 2 mm	Less Than 2 mm	Less Than 100 µm	Less Than 10 µm	Less Than 2 mm	Less Than 100 µm	Less Than 10 µm[a]
Trinity (g)	341	1147	578	34	1.0	0.50	0.06
(pCi/g)	-	1.3 ± 0.2[b]	1.9 ± 0.10	2.0 ± 0.10		1.5	1.5
(pCi)	-	1468	1087	138[a]		0.74	0.09
				(68)			
Rocky Flats							
1A (grams)	133	502	188	46		0.38	0.2
(pCi/g)	-	635 ± 91	1580 ± 160	1680 ± 160		2.5	2.6
(pCi)	-	3.19 E5	2.96 E5	1.69 E5[a]		0.93	0.5
				(7.73 E4)[d]			(0.24)[d]
1B (grams)	140	487	178	44		0.37	0.2
(pCi/g)	-	634 ± 94	1050 ± 130	1730 ± 190		1.6	2.7
(pCi)	-	3.09 E5	1.87 E5	1.69 E5[a]		0.61	0.6
				(7.61 E4)[d]			(0.25)[d]
1C (grams)	169	568	203	20		0.36	0.2
(pCi/g)	-	593 ± 83	939 ± 113	1460 ± 160		1.6	2.5
(pCi)	-	3.37 E5	1.91 E5	1.66 E5[a]		0.57	0.5
				(2.92 E4)			(0.087)[d]
Rocky Flats							
2A (grams)	93	591	319	71		0.54	0.2
(pCi/g)	-	642 ± 95	1030 ± 120	1590 ± 150		1.6	2.0
(pCi)	-	3.79 E5	3.30 E5	1.88 E5[a]		0.87	0.5
				(1.13 E5)[d]			(0.30)[d]
2B (grams)	88	462	213	40		0.46	0.2
	Samples not analyzed						
2C (grams)	72	490	216	38		0.44	0.2
(pCi/g)		838 ± 142	Other samples not analyzed				
Rocky Flats Average (grams)					1.0	0.43 ± 0.07	0.2[a]
(pCi/g)		670 ± 100[c]	1150 ± 300	1620 ± 120	1.0	1.8 ± 0.5	2.4 ± 0.3
(pCi)					1.0	0.75 ± 0.18	0.5

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[a] Six percent of the Trinity soil mass and 20 percent of the Rocky Flats soil mass were assumed to be in this size.

[b] Two sigma counting errors.

[c] Standard deviation.

[d] The value in parenthesis is the actual quantity of picocuries or fraction thereof recovered in the less than 10 µm fraction. See footnote a and item 6 on page 42 of the text.

En indicates 10^m; e.g. $E5=10^5$

TABLE 14. SOIL SIZE MASS AND ACTIVITY FRACTIONS OF VARIOUS INVESTIGATORS

Investigator	Sample Location	Fraction of Total Sample Greater than 2 mm	Ratio of the sample mass and plutonium concentration in the less than 2 mm fraction which was in the less than:**				Depth of sample	Ultrasonic Dispersion Used	Remarks
			100 µm fraction		10 µm fraction				
		(mass basic)	(mass)	(activity)	(mass)	(activity)	(cm)		
ORP-LVF	Trinity	0.23	0.50	1.5	0.05	1.5	2.5	No	
Tamura (1975)	NTS #1	0.20	0.34	--	0.033	--	5	No	Desert Pavement
	#1	0.20	0.40	--	0.083	--	5	Yes	Desert Pavement
	#2	0.027	0.45	--	0.011	--	5	No	Desert Mound
	#2	0.027	0.51	--	0.086	--	5	Yes	Desert Mound
ORP-LVF	Rocky Flats		0.43±0.07	1.8 ± 0.5	0.2 ± 0.1	2.4 ± 0.3	1	No	4 Samples
Johnson et. al.	RFP (4 samples)	--	--	--	0.28±0.12	5 ± 50%*	surface	Yes	< 17 µm, 1g/cm^3, used*
	(4 samples)	--	--	--	0.25±0.04	3 ± 25%*	dust	Yes	H_2O_2 to break bonds
	(7 samples)	--	--	--	0.36±0.09	2 ± 40%*	surface	Yes	Dry sieved, after drying
	RFP (1 Bkgd.)	--	--	--	0.49	~6 *	dust	Yes	
Little et. al. (1973)	Rocky Flats	0.31	0.05	1.8	--	--	3		Dry sieved
	Rocky Flats	0.40	0.13	~ 4	--	--	3		Dry sieved
Tamura (1975)	ORNL	0	1	--	0.30	--	7.5	No	Flood plain silt
		0	1	--	0.38	--	7.5	Yes	Flood plain silt
Tamura (1975)	Mound Lab	0	0.87	--	0.30	--	Core	No	Silt
	Ohio	0	0.91	--	0.37	--	Core	Yes	Silt

* The Plutonium concentration in the total sample is not based on the sample of material. Rather it is based on Johnson's et. al. adaption of isopleths from Krey and Hardy (1970).

The values were estimated by dividing the average plutonium in soil concentrations, for given areas, for particles less than 5 micrometers in diameter (density 11 g/cm^3) from Johnson et al. by the plutonium concentration in the total soil estimated from isopleths that Johnson et al. adopted from Krey and Hardy (1970). The error term is the standard deviation from averaging the results.

The reference to a 17 um diameter relates to the equivalent aerodynamic diameter in air.

**Mass refers to the mass fraction of material (i.e., g/g).

Activity refers to the ratio of plutonium concentrations (i.e., pCi/g:pCi/g).

destroy any organically-bonded conglomerates, and ultrasonic mixing to further destroy conglomerate bonds. The intent of the ORP-LVF treatment was to preserve the basic conglomerates that would not disperse in a water suspension.

The results of Little et al. (1973) indicate a relatively small fraction of material in the less than 100-micrometer size class. This may be due to the difference in samples, or to oven-drying the sample (which stablizes the conglomerates) and dry sieving versus wet sieving the sample at 140 mesh.

The results of Tamura (1975) from Mound Laboratory, Ohio and Oak Ridge National Laboratory (ORNL) are for silt samples. Thus, it was expected that a large fraction of the material would pass 140 mesh.

AIR SAMPLING TECHNIQUES

Inhalation is the primary human intake pathway for plutonium; thus air sampling data are the preferred environmental data for inhalation exposure evaluation. In order to assess inhalation hazards from soil sampling, it is necessary to postulate factors for resuspension and atmospheric transport of plutonium. Air sampling provides direct evaluation of atmospheric concentrations of plutonium from airborne releases prior to deposition, and a direct measure of resuspended material. Furthermore, soil samples only provide results for discrete points within potentially heterogeneous areas--whereas air sample results indicate the average concentration for plutonium over a general area.

Air sample results are not always generally applicable to human exposure or even to actual atmospheric concentrations of the sampled material. For plutonium, the emphasis is on the particulate material. Thus, there are the concerns of:

a. Isokinetic sampling--sampling at the air stream flowrate so that the particle size distribution of the sample is representative of that in the atmosphere.

b. The air sampler face velocity or linear flowrate should be representative of human biophysical parameters. If the sampler inlet configuration and linear flowrate are not properly designed, the sampler will not obtain a sample of the representative particle-size distribution inhaled by man. Intake and deposition within the respiratory tract is dependent on the equivalent aerodynamic particle-size distribution of the inhaled material.

c. If the sampler is of the filtration type, the filtration material must be such as to provide retention of the airborne material at the sampling flow-rate. The dust-loading pressure drop characteristics of the filtering material must be considered also.

d. The sampler must be properly located so that it obtains a representative sample of the atmosphere, i.e., not on the leeward side of buildings or hills.

Given the above comments, it becomes apparent that a random air sample does not necessarily provide all necessary hazard-assessment information. Sampling parameters must be defined.

There are several reference works on aerosol-sampling technology (Mercer, 1973). Thus, this section will not attempt to discuss the physics of aerosols. This section will focus on aerosol-sampling techniques pertinent to assessing airborne concentrations of plutonium. The emphasis will be on techniques designed for adequate flowrates and sampling times for assessing environmental concentrations (less than 0.1 fCi of plutonium per cubic meter of air). A minimum detection level of about 20 fCi is required on the final separated sample. Assuming a chemical yield of 50 percent, and given the uncertainties in sample analysis, a reasonable minimum required activity in the sample is 80 fCi. The necessary sampling rate is equal to the minimum required sample activity divided by the product of air concentration and sampling time. Thus, assuming an air concentration of 0.05 fCi/m^3, a sample volume of 1600 cubic meters is necessary. Therefore, for a flow-rate of one cubic meter per minute, the sampling time required is 1600 minutes, or about one day.

Work by Bagnold (1954) and Chepil and Woodruff (1963), referenced by Anspaugh et al. (1975) and Buck et al. (in press), indicates that saltation and surface creep account for the majority of airborne soil movement. These processes generally include soil particles from 50-500 micrometers and 500-2000 micrometers, respectively. Thus, although these processes generally move particles near the ground (within one meter), air samples should be scrutinized to insure that they do not contain large amounts of material above the respirable particle-size range (5 to 10 micrometers).

The phenomona of resuspension is generally related to particles ranging up to 50 micrometers. Thus, only a fraction of windborne material related to resuspension is respirable, and resuspension accounts for less than 10 percent of airborne soil movement.

PHYSICAL CHARACTERISTICS OF AEROSOLS

The physical characteristics of airborne particles are generally described by their aerodynamic characteristics. In simple terms, the forces acting on a particle are proportional to the density of the particle and the square of the diameter of the particle for particles of the density and diameter of interest for plutonium inhalation (Mercer, 1973; Morrow, 1966; and ICRP, 1972).

Deposition of plutonium-related particles greater than 10-micrometer aerodynamic diameter in the pulmonary section of the lungs is essentially zero (Mercer, 1973 and Morrow, 1966). The aerodynamic diameter relates to the equivalent diameter of a particle with a density of 1 g/cm^3 which responds similarly in air streams to the subject particle.

The terminal setting velocity of a particle can be described as (Eisenbud, 1963):

$$V = 0.003\ \rho d^2$$

Where V = velocity (cm/sec)

ρ = density of particle (g/cm^3)

d = particle diameter (μm)

This equation is applicable for particles with streamline motion (e.g., density less than 10 g/cm^3 and diameter less than 50 μm or d less than $115/\rho^{1/3}$). When the diameter of a particle is less than the mean free path of gas molecules, Stokes' equation underestimates the terminal settling velocity. This can be corrected for by using Cunningham's modification of Stokes' equation (Eisenbud, 1963):

$$V_c = V_s[1 + (1.7\lambda/10^{-4}d)]$$

Where V_c = corrected velocity

V_s = Stokes' Law velocity

λ = mean free path of gas molecules,

about 10^{-5} cm at sea level.

d = particle diameter, μm

The air entering the nose is actually deficient, with respect to the ambient air, in particles having settling velocities similar to the inhalation face velocity and normal wind speeds. (ICRP, 1966). The inhalation face velocity is (ICRP, 1966):

$$1500\ \text{ml} \times \frac{15}{\text{min}} \times \frac{\text{nose}}{1.5\ \text{cm}^2} \times \frac{\text{meter}}{100\ \text{cm}} \times \frac{\text{min}}{60\ \text{sec}} = 2.5\ \text{m/sec}$$

The assumptions are:

a. Tidal or inspirational volume, 1500 ml

b. Inspiration rate, 15 per minute

c. Cross sectional area of nostril, 0.75 cm^2 or 1.5 cm^2 for nose.

The above parameters correspond to a reasonable level of activity, somewhat equivalent to industrial workers. Basal metabolism is about 500 ml tidal volume with 12 respirations per minute (Comroe et al., 1963). Mild to moderate activity would relate to 750 cm^3 and a respiration-rate of 15 per minute (ICRP, 1966).

Air sampler face velocities vary over a wide range. A volume of 1 l/min for a 1 cm diameter filter (0.785 cm^2) relates to a velocity of 0.21 m/sec (1 ft^3 /min through a 1- in diameter filter is 0.93 m/sec). Thus, for a nominal high volume sampler (1 m^3/min for an 8 by 10 inch filter having an effective filter area of 7 by 9 inches), the face velocity is 0.41 m/sec. If a 4 in-diameter filter was used, the velocity would be about 2 m/sec.

A nominal wind velocity of 10 miles per hour is 4.47 m/sec. Thus, it becomes apparent that it is difficult to sample isokinetically with conventional filter-type samplers. The high volume sampler only has a face velocity of 2 m/sec with a 4 in-diameter filter. Even the face velocity for the human nose is about 2.5 m/sec, or the equivalent of 5 miles per hour.

Patty (1958) indicates that the air velocity drops to about 10 percent at one diameter from the face of an exhaust vent. Thus, given the ratio of the diameters of air samplers and human nostrils (generally 10 cm versus less than 1 cm, respectively), air samplers with face velocities 0.25 m/sec generally should be equivalent to the human nose. Furthermore, the settling velocity of a 10 μm aerodynamic equivalent particle is only about 0.3 cm/sec; i.e., two orders of magnitude less than a sampler face velocity of 0.25 m/sec.

The above does not resolve the problem of subisokinetic sampling rates. If the sample face is oriented downwind, there is a definite probability of unrepresentative sampling due to an inadequate capture velocity. Sehmel (1973) has generally resolved this problem by placing samplers on a pivot. The sampler orientation is then controlled by a wind-oriented cowl, so that the sampler is oriented into the wind.

TYPES OF AIR SAMPLERS

There are several techniques for obtaining samples of the particulate material suspended in air. They include:

1. Mass air samplers where the air stream is drawn through a filter medium.
2. Electrostatic precipitators where the particulates are removed from the air stream by electrostatic force.

3. Impactors which normally are used to segregate the particulates into various size categories. Particulates are impacted on various stages (size categories) as a result of channeling the air stream around the impaction plane.

4. Air elutriation sampling techniques separate particles on the basis of the settling velocity. The differential of the settling velocity and the velocity of the air current in which the particle is moving is used to separate particles based on their size and density.

5. Aerosol centrifuges utilize the same principle as elutriators, except centrifugal force is used in place of gravitational force.

Many samplers utilize several of the techniques. Andersen impactors use several impactor stages to segregate particles into size fractions from 1 or 2 to 10 micrometers and a filter to collect the smaller particles. The sampler may be designed to exclude material over 10 micrometers. Some characteristics of the various types of samplers are discussed below.

Mass or Filter-type Air Sampler

An air mover is used to draw air through a fibrous or membrane-type filter. Although particle sizing can be done either through optical or audioradiographic techniques, or by using filter packs containing filters with different size penetration characteristics, normally particle sizing is not done for filter-type samples.

Filter-type samplers come in a large range of sizes; from personnel monitoring devices having flowrates of liters per minute, to the high volume samplers at about 1000 liters per minute. Anspaugh et al. (1974) report on an ultra high volume sampler capable of flowrates of 25,000 liters per minute. This sampler was designed to obtain samples of resuspended dust over short periods of time.

The principal parameters for filtration-type samplers are the flowrate, face velocity, and the filtering medium. The flowrate determines the volume of air sampled per unit time and, thus, in part, dictates the sampling time, assuming the amount of material collected is near the MDA.* The face velocity affects not only the characteristics of the aerosol drawn into the sampler, but also the fraction of material collected by the filter. The decision concerning filtering material must be based on face velocity, expected dust loading, proposed analytical techniques, particle size retention requirements, and pressure drop characteristics.

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* MDA; minimum detectable activity, see Sample Analysis Techniques section.

Electrostatic Precipitation

This is a two-stage process. First, the particles must be charged in a unipolar ion field. In the second stage, a strong electric field is used to precipitate the charged particles on a suitable collection surface (Mercer, 1973). Collection efficiencies of 99.9 percent can be obtained for particles of 0.2 to 0.7 micrometer mass median diameter; whereas particles around 0.2 micrometer are difficult to collect because of the small charge retained by the particle. Because of design complexities and power requirements, electrostatic precipitators are not used commonly in environmental sampling.

Recently, the U.S. Environmental Protection Agency laboratory at the Research Triangle Park in North Carolina has participated in the development of an ultra high volume sampling system. The system is capable of flow-rates of 26 m^3/min. The particulate material is segregated into respirable and non-respirable material by use of an electrostatic precipitator and other techniques.

High-Volume Cascade Impactors

Cascade impactors are composed of a series of impactor stages and a final filter. The units of interest generally provide several fractional steps for particles between 1 and 10 micrometers in diameter, with the final filter collecting material less than 1 micrometer in diameter. Cascade impactors have demonstrated their ability to provide particle-size distributions, based on the equivalent aerodynamic diameter, for ambient levels of airborne particulate material.

A single impaction stage is composed of a plate with precision-machined orifices followed by an impaction plate. The impaction plate contains the orifices for the next stage. The airstream flows through the orifice, and as it is impinged on the impaction plate the airstream splits to go on through the adjoining orifices. The inertial qualities of the particles cause those in the designed size spectrum to be impacted on the impaction plate, directly below the orifice. The deflected airstream goes through the adjoining orifices. The orifices are of a slightly smaller diameter than the previous stage. Thus, the constant air volume, but smaller orifice diameter results in an increased air velocity, with the resulting impaction of the next size smaller particles on the following impaction plate.

Mercer (1973) presents a detailed account of the theory of impaction units. There is not a discrete cutoff of particle size increments with each stage. Mercer (1973, Figure 6.36), illustrates the general fractionation that occurs. The hypothetical aerosol is assumed to be made up of unit density particles with a geometric mean diameter of 10 micrometers and a geometric standard deviation of 2. The effective cutoff aerodynamic diameters (ECAD) are 16, 8, 4, and 2 micrometers.

It generally is assumed that all particles collected on a given stage have aerodynamic diameters larger than the ECAD for that stage. A stage not only does not collect all particles above the ECAD, but collects some particles smaller than the ECAD. Some material is assumed to be collected that is not collected, and some material assumed to be passed by a stage is actually retained. These are about equal for each stage. Thus, the actual mass per stage is approximately correct. The differences in the mass for a stage generally are less for round jets than for rectangular jets (Mercer, 1973, p.234). The ECAD is the diameter for a particle which has a 50 percent probability of retention on the subject stage.

In addition to errors resulting from non-ideal design there are several potential sources of error. These include wall loss of material, disaggregation of particles, and rebound and re-entrainment of deposited material.

Wall losses refer to the retention of impacted material in the impaction stages other than at the intended impaction area. Mercer (1973, p.235) reports wall losses ranging from 14 percent to 2 percent for high sample volumes for a low volume sampler (i.e., 0.05 to 0.15 l/min).

Wall losses result from non-laminar flow between the stages. However, the wall losses can be extenuated by rebound and/or re-entrainment of the impacted material. Mercer(1973, p.236) notes rebound is a serious problem if the collection surfaces are not coated with a soft layer to cushion the impact of particles (e.g., if the impactor plate is used as the collection medium versus using a filter for collecting the impacted material). He also notes that both rebound and re-entrainment put an upper limit on the amount of material that can be collected on a stage without degrading the operation of the instrument.

Sehmel (1973) defines wall loss as the amount of material associated with the walls directly above the stage of interest, divided by the amount of activity on the stage of interest. He reports wall losses for the Andersen 2000, Inc., Model 65-100, 20 ft^3/min, high volume unit for stage loadings between about 50 and 200 mg. The losses vary from about 1 percent at 50 mg/stage to 5 to 20 percent at 200 mg/stage.

Sehmel (1974) provided additional information on the wall losses for the Andersen 2000 Inc., unit. He noted the following:

1. There appears to be no direct relationship between interstage losses and stage loading.

2. The data indicate wall losses of up to 20 percent. Operations during dust storms would undoubtedly result in higher wall losses.

3. The average particle size in the interstage material (wall loss) for each stage was much larger than should have been present for the respective stages. Apparently, some nonrespirable particles work their way through the various stages. This tends to give results that are conservative (i.e., there is more respirable material indicated as being present than actually is present in air).

There appear to be only two high-volume impactors available through commercial sources. The Andersen 2000 Inc. is based on 20 ft^3/min flow rate (566 l/min). The unit has four stages with cutoffs at 7.0, 3.3 , 2 , and 1.1 micrometers, with a backup filter for material less than 1.1 micrometers (Burton et al., 1973). The unit is about 30 cm in diameter and can be matched to high-volume air samplers. The operation of the unit has been reviewed by Burton et al. (1973) and Sehmel (1973, 1974). In addition to the previously indicated information, Burton et al. (1973) note that some types of fiberglass filters are prone to absorb atmospheric acid gases. Thus, the total mass amount of collected material cannot be directly related to a mass air sample result for a single sample. Apparently, fiberglass filters with a pH adjusted to 6.5 largely resolve the problem.

Tech Ecology, Inc. markets a 5-stage cascade impactor designed for a flow rate of 40 ft^3/min (1,130 l/min). Tech Ecology model 252 has size cutoffs of 8.2, 3.5, 2.1, 1.0, and 0.5 micrometers, with a final filter for less than 0.5 micrometers. The unit is rectangular and fits the standard 8 x 10-inch high-volume filter holder. The impactor orifices are rectangular slits 12.5 cm long. An advantage of the design is the small amount of filter paper (about 170 cm^2) that has to be analyzed for results from each stage. There appear to be no published reports evaluating this unit.

Sehmel (1973) reports results of a study with the impactor facing into the wind, and with the impactor face pointed vertically up or down. About 50 percent more material was collected with the sampler pointed up versus down. The results with the sampler oriented into the wind, with a wind directed cowl, fell between the upward and downward oriented sampler, and were considered to be the most valid of the three sets of results. The data were obtained using Andersen 2000 Inc. samplers. The flowrate was 20 ft^3/min (570 l/min), and the linear velocity for the 6-in (15-cm) diameter cowl was 0.54 m/sec (1.2 miles/hr).

Air Elutriator and Centrifugal or Cyclone Samplers

Air elutriators and cyclones utilize similar processes. The settling velocity of the particle is used to fractionate material in elutriators, whereas cyclones utilize centrifugal force concepts. Both systems generally are used in two-stage samplers. The elutriator and cyclone stages are used to remove the non-respirable material from the air stream. The respirable material (less than several micrometers in diameter) is collected on a filter in the second stage.

The fractionated particulate material passed by the cyclone generally relates to the definition of respirable material designated as the Los Alamos Scientific Laboratory (LASL) criterion. The horizontal elutriator passes material which relates to the criteria of the British Medical Research Council (1961). The LASL criteria resulted from a meeting called by the Atomic Energy Commission, Office of Health and Safety, at Los Alamos in 1961. Thus, the term AEC criteria is also used. The American Conference of Governmental Industrial Hygienists set forth a slightly revised version of the LASL criteria (Federal Register, 1969). These criteria, are summarized by AIHA (1970) and Ettinger et al. (1970).

Air elutriation is a process of particle separation based on the settling velocity of the particle. This process may be done on either a horizontal or vertical plane--thus horizontal or vertical elutriators. Both techniques are based on the comparison of particle settling velocities and the velocity of the air stream transporting the particle.

In vertical elutriators, the particles are carried upward in a diverging air stream until they reach a point in the air stream at which their settling velocity equals the vertical component of the diminishing air velocity. Vertical elutriators have been used for size-fractionation of powders, but have received little use as air samplers (Mercer, 1973, p. 192).

In horizontal elutriators (HE), the particle settling velocity is normal to the transporting air velocity. The air stream passes through a horizontal duct. The distance from the duct inlet at which particles fall out is inversely proportional to their settling velocity or aerodynamic particle size. The size distribution of material along the path length of the duct varies, thus indicating the potential for obtaining an indication of the size spectrum.

The vertical dimension of the inlet air duct of a HE is generally a significant fraction of the total vertical fall height. Thus, particles entering at the lower level of the duct have a reduced fall height, compared to particles entering the upper part of the duct. Thus, there is a general spread of the size distribution along the horizontal length of the HE. Mercer (1973) indicates that a sharp cutoff can be obtained with a properly designed HE. Wright (1954) indicates the comparison of the actual and theoretical retention values for the 100 l/min Hexhlet instrument. The following retention values generally are obtainable (Wright, 1954; AIHA, 1970; and Mercer, 1973):

Particle Size (micrometers)	Percent Retention (In HE)
1	∿2
<2	10
5	50
7	100

Lippmann (1970) describes various HE's. Although most of the units operate in the liter-per-minute category, Wright (1954) presents data on the Hexhlet unit, designed for 100 l/min. The design was subsequently revised to 50 l/min. Shanty and Hemeon (1963) discuss a unit designed for a flowrate of 1250 l/min.

Lippmann (1970) notes it is generally difficult to collect the material from HE units for analysis. In many designs, it apparently is difficult to clean the HE adequately, to prevent future samples from being contaminated by re-entrained material. A preference for cyclone separators is noted.

Centrifugal or cyclone samplers (CS) separate particles based on their centrifugal force (i.e., mass and diameter, or equivalent aerodynamic diameter). They are more flexible than HE's in that they can be operated in any position. Thus, small cyclones have been developed as personnel monitors. Lippmann (1970) indicates a listing of CS's, most of which are in the liter-per-minute flowrate range, although one unit with a turbine blower is rated at about 1000 l/min. Volchok et al. (1972) report results from the Rocky Flats, Colorado area using a 100 l/min CS described by Lippmann and Harris (1962).

The design parameters on a CS are critical. Ettinger et al. (1970, Table 4) indicates the change in the cyclone retention with flowrate for a one-half inch unit. Lippmann (1970) reports that most of the cyclone calibrations prior to about 1970 were in error. The errors were due to an overestimate of particle sizes, as a result of the microscopic measurement technique used. Apparently the disagreements range up to a factor of two in flowrate for describing a given size cutoff. Given this, the data presented by Ettinger et al. (1970, Table 4) would indicate roughly up to a factor of two error in cyclone retention for 2-micrometer particles.

Combination Electrostatic Precipitation and Cascade Impaction

Henry and Mitchell (1975) present data on a high-volume sampler developed for EPA, Research Triangle Park, North Carolina. The sampler is designed for 28 m^3/min (1000 ft^3/min). It contains impaction stages designed for particles greater than 3.5 micrometer and 1.7 to 3.5 micrometer. The final stage, for less than 1.7 micrometer particles, is an electrostatic precipitator. There does not appear to be any published information evaluating the operation of the unit.

TYPES OF FILTRATION MATERIAL

Many of the characteristics and limitations of filtration samplers relate to the filter medium. Based on their physical structure, filters can be classified as either fibrous mats or porous membranes. Filters have varying particle size retention characteristics, and the characteristics of a given filter are dependent on the airstream face velocity. Other considerations include dust loading and associated pressure drop, and the presence of trace materials (e.g., uranium, thorium, and radium) in the filter material.

The theory of fibrous mats is discussed by Mercer (1973, p.115). Fibrous filters are made of cellulose fibers, plastic fibers, glass fibers, and other materials including asbestos. The filter performance is closely related to the diameter of the fibers, with the smaller diameter fibers having better collection properties. Collection of particles on filters is not solely a sieving phenomena; rather, it is due to electrostatic forces, interception, impaction, and diffusion.

Most common filters, fibrous or membrane, have adequate particle collection efficiencies for air sampling; however, it has been noted that Whatman 41 cellulose fiber filters have a fairly low efficiency (70 to 80 percent) at low face velocities of 20 to 30 ft/min (about 0.13 m/sec or 0.28 mi/hr). This is equivalent to a flowrate of about 2 ft^3/min (60 l/min) through a 4-inch (10-cm) diameter filter.

Unpublished information from a study by Eadie, ORP-LVF provides data on the dust loading and pressure drop properties of several filters. Tests were conducted on 4-in (10-cm) diameter Whatman 541, Acropor, Gelman Type E Glass Fiber, and Microsorban filters at initial flow rates of about 10 ft^3/min (280 l/min). The results indicate that the glass fiber and Microsorban filters had better dust loading properties than the other filters. Glass fiber filters showed a 30 percent decrease in flow rate with a filter load of 260 mg. Microsorban indicated less than a 10 percent decrease in flow rate with a load of 200 mg, the highest load used on the Microsorban tests. Conversely, Whatman 541 paper indicated a 60 percent flow rate decrease with a dust load of 200 mg or less.

The filter composition affects the difficulty and precision of radiochemistry analysis. The ease of wet or dry ashing Whatman paper always has made it a favorite with chemists. The difficulty of dissolving fiberglass filters and the associated trace elements put in solution have provided difficulty for radiochemical analysis. Even with refined techniques, the EPA's Las Vegas analytical laboratory has found about 80 in^2 (500 cm^2) to be the maximum amount of fiberglass material to be amenable to plutonium chemistry analysis.

Microsorban, a polystyrene fibrous mat material, is very amenable to radiochemistry. If dried and heated for several hours at increments of 100°C to 350°C, it can be white-ashed to a powder at 600°C. When put in solution, it essentially has zero residual (Golchert, Argonne National Laboratory, Personal communication, Feb. 1975).

Filtering materials contain numerous trace elements. These elements include uranium and thorium progeny, and many metals (especially in the fiberglass filters). The amounts and variances of the trace elements significantly effect the sensitivity of monitoring low levels of these trace elements in air. Table 15 indicates values determined by ORP-LVF for some of these contaminants in several filters.

The analyses for many of the radionuclides are incomplete. However, it is evident that most of the filter materials contain varying amounts of radium-226, uranium and thorium. Admittedly, some of the variation may be due to analytical or counting error, but many of the results were based on a composite of four filters.

Golchert, in a private communication on Feb. 11, 1976, noted that Argonne National Laboratory has detected concentrations of 4 to 18 fCi of thorium-232 and 2 to 8 fCi of uranium-238 per 780 cm^2 of Microsorban. These relate to average values of about 1 fCi and 0.5 fCi, respectively of thorium-232 and uranium-238 (1 fCi total uranium) for a 4-inch diameter (10-cm) Microsorban filter. These values are significantly lower than the radium-226 values given in Table 15.

Given several assumptions, these trace contaminants can be related to equivalent air concentrations. Assuming a sampled volume of 2000 m^3 (1.4 m^3/min for 1 day), a contamination level of 0.2 pCi/filter is equivalent to an air concentration of 0.1 fCi/m^3. This is about one-fifth of the nominal radium-226 ambient concentrations (see section on natural activity).

TABLE 15. RADIONUCLIDE LEVELS IN AIR FILTERS (ORP-LVF)

				pCi per Filter							
Filter Material	Number of Samples	Weight Ave. (g)	Weight Range	Ra-226 Ave	Ra-226 Range	U-238 Ave	U-238 Range	Th-230 Ave	Th-230 Range	Th-232 Ave	Th-232 Range
Whatman 41	4	0.62	1.06	0.16	1.82						
Whatman 541	2	0.62	1.01	0.30	1.28						
Gelman Glass Fiber	7	0.52	1.04	0.26	1.96	0.085	6.15	0.18	1.63	0.013	2.00
Microsorban	5	1.47	1.05	0.17	3.73						
Acropore	5	0.41	1.02	0.90	5.7						
Millipore	1	1.7	----	0.1	---	<0.01					

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* The range is the ratio of the highest result to the lowest result.

There appears to be minimal, if any, plutonium contamination in air filter materials. Thus, these contaminants are somewhat academic for sampling related solely to plutonium. But their presence should be recognized in determining methods for plutonium analysis and when considering gross alpha measurements.

The variance of filter weights has to be recognized if the specific activity of the material on air samples is to be determined. Mercer (1973) notes that cellulose fiber filters are prone to collect moisture from humid air. At 100 percent relative humidity, a cellulose filter may gain 17 percent weight, compared to its dry weight, versus 0.1 percent for fiberglass filters.

AMBIENT CONCENTRATIONS OF NATURALLY-OCCURRING ALPHA EMITTERS

The ambient concentration of plutonium-239 in air is roughly 30 aCi/m^3. This is significantly below the standard concentration guide of 10 CFR 20 for individuals in the general population which is 60,000 aCi/m^3.

Ambient concentrations of the naturally- occurring alpha emitters range over several orders of magnitude. Values vary from yearly averages of 100 aCi/m^3 of total uranium, about 30 aCi/m^3 of thorium-238 and 232, and 50 aCi/m^3 of thorium-230 (AEC, 1974a) to 2000 aCi/m^3 of polonium-210 (AEC,'1973a).

Based on information in the previous section, roughly 100 aCi/m^3 of the various nuclides could be accounted for by contamination in the filter material, depending on the filter material used and volume of air sampled per quantity of filter material.

By way of illustration, the total gross alpha activity on an air filter result could be roughly 0.9 pCi under the following conditions . Assuming a sampling rate of 1 m^3/min, a sampling time of 100 minutes, and an estimated nominal ambient background of 4000 aCi/m^3, the gross alpha value is comprised of 0.5 pCi from natural contaminants in the filter and 0.4 pCi of activity collected during the sampling period.

The gross alpha estimate is somewhat greater than the average gross alpha estimates from Argonne National Laboratory (ANL), Illinois, 2,500 aCi/m^3 (AEC, 1974a); Rocky Flats, Colorado, 5,000 aCi/m^3 (AEC, 1973a); and Los Alamos, New Mexico, 1,000 aCi/m^3 (AEC, 1973a). The above gross alpha results are based on longer run times (days); thus, the filter contamination becomes less significant (estimated at 5,000 aCi/m^3 in our hypothesized value). Also, the ANL and Los Alamos results are based on Microsorban filter material, which has a contamination value lower than the postulated value. In addition to these factors, the hypothesis of the gross alpha air concentration was based on higher-than-normal values of natural radionuclides in the atmosphere. Even with the noted conservative assumptions, the postulated ambient gross alpha estimate of 9,000 aCi/m^3 is significantly below the plutonium-239 concentration guide of 60,000 aCi/m^3.

ANALYSIS OF AIR SAMPLES

Analysis of air samples generally is equivalent to the analysis of soil samples, plus considerations of the sampling medium if a filter is used. The medium generally does not present unusual problems, except in the case of fiberglass filters. Most membrane filters, Microsorban, and cellulose fiber filters generally can be wet or dry ashed to a low residual. The specific activity of naturally occurring uranium and thorium radionuclides in air samples generally is similar to their specific activity in soil (Golchert, ANL, personal communication, February 1976).

Plutonium in air samples stems from both resuspended soil and fallout. In areas with an air concentration of about 30 aCi/m^3 from atmospheric fallout and deposition on the soil of less than 10 - 30 nCi/m^2 (i.e., about 10 times background), the plutonium concentration in air is largely a result of atmospheric fallout.[a] Thus, the particulate material on air filters has a higher specific activity than that in soil. The information on sample analysis in the following section is applicable to air samples.

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[a] Douglas, ORP-LVF personal communication, February 1976 and A. Hazle, Colorado State Department of Health, personal communication, February 1976.

G. Merrill (Air Force McClellan Central Laboratory, verbal communication, May 3, 1976) indicated that using plutonium isotopic ratios from mass spectrometry, a contribution from resuspended Trinity contamination (up to tens of percent) could be detected in the data from Douglas.

SAMPLE ANALYSIS TECHNIQUES

In many situations it appears that the analysis of samples is not integrated with the philosophy of collection of the samples and the objectives of the overall program. Plutonium in soil, as well as in other media such as animal tissue, exemplifies this situation, because of its potentially heterogeneous distribution. The objectives of the program may dictate compositing up to ten discrete soil samples, to insure a sample representative of the sampled location. The total sample, composed of several kilograms, may be milled and mixed, with only a small aliquot taken for the actual analysis. This aliquot may vary from as small as one gram (EMSL-EPA, Las Vegas prior to January 1975) to about 100 grams (Krey and Hardy, 1970). The aliquot size for analysis is related to the difficulties of dissolution of large quantities of soil, and for fusion techniques the limitations and costs of the required analytical apparatus.

In most instances, the analyst follows the philosophy of taking an aliquot which he thinks can be adequately analyzed. The potential presence of discrete particulate plutonium in the sample, and the probability of obtaining a representative fraction of the material in the aliquot, may not be addressed.

The problem of adequate sample size also relates to some biological samples, such as bovine livers, kidneys, and bones, etc., where it may not be convenient to analyze the whole sample. Consideration of the heterogeneous structure of organs is necessary if analyses of aliquots of the organs are to be meaningful.

ANALYTICAL SENSITIVITY

Analytical sensitivities are generally related to the counting error (Johns, 1975; Sill, 1971; Krey and Hardy, 1970; Chu, 1971; and Eberline, 1974). In many instances, the minimum detectable activity (MDA) is defined as a value which is equal to the 2-standard deviation (SD) or 95 percent confidence level (C.L.) value (e.g., 20 fCi ± 20 fCi). Such results are normally presented as less than values (e.g., <20 fCi).

The use of a value of less than the 2-SD value results in the significant probability of an erroneous statement. If one believes in the validity of the counting error, there is only a 50 percent probability that the value is less than the 2-SD value (e.g., <20 fCi). In order to have a 95 percent confidence level statement, a value of the mean plus 2-SD should be used.

Eberline Instrument Corporation (1974) uses another fairly standard technique of three times the background counting error. This gives an MDA somewhat less than the 2-SD equal to the mean technique. But, as in the 2-SD technique the statement of a less than value has a 50 percent or greater probability of being in error.

Robinson et al. (1975) considered the range of background samples for plutonium-238 and the variation of results for low concentrations. The objective of the study was to assess the inventory above the baseline or background level near the Mound Laboratory, Miamisburg, Ohio. Aliquots of a sediment sample from 50 miles upwind of the plant were used for background determinations. The reported gross concentrations in the background samples (no system background subtracted) ranged from 0.000 to 0.765 pCi/g with a mean of 0.077 ± 0.040 (1-SD for 50 values). The minimum detectable level was set at 0.1 pCi/g. Using 0.1 pCi/g and recognizing that background values ranged up to 0.8 pCi/g, the sample results were reported as less than 0.1 pCi/g or the actual result for values above 0.1 pCi/g. Blank background values were not subtracted from the results. Actual plutonium-238 background values for this area were reported to be 0.0002 pCi/g for 30 cm (12 in.) depth cores, or roughly 0.002 pCi/g for the top 5 cm (2 in.).

Although the results of Robinson et al. are not directly applicable to studies at background levels, the concept of using the variation in low level results, versus the counting error, to define the MDA has merit.

The sensitivity of analytical procedures is inherently a function of five parameters, some of which are reasonably fixed, but several of which can be varied. The parameters are:

1. Sample size: The sensitivity depends on the total amount of activity present. Thus, ideally the sensitivity of a 10-gram sample is one-tenth of that for a 1-gram sample. The acid dissolution and fusion techniques tend to have a nominal maximum of about 10 grams of sample. The ease of analysis, size of vessels and quantities of interfering elements generally result in the analysist's preference for a sample smaller than 10 grams. The size refers to the dry weight of soil, or weight of ash for biological samples.

2. Radiochemical yield: The yield is not an independent variable. Mullins (EMSL-LV, verbal communication, Feb., 1975) noted that although yields of 90 percent plus were obtainable with 1-gram soil samples, the yield for 10-gram samples had been about 50 percent,

although there was hope for improving it. The drop in yield is due to the interference from the increased quantities of elements such as calcium and iron. Thus, the 10-gram samples are only the equivalent of 5-gram samples, or less, but there is still the benefit of obtaining a more representative aliquot. There is an additional uncertainty with low yields, due to the uncertainty in the yield determination. A measured recovered activity divided by a yield of 90 percent (with an uncertainty of 10 percent) has a much lower uncertainty than a value divided by 50 ± 10% or 20 ± 10%. There is the additional uncertainty related to the conventional propagation of error techniques (Parrott, 1966 and Pugh and Winslow, 1966). The simple technique for the square root of the sum of the squares of the coefficient of variation only applies for the division of parameters if the coefficient of variation is at most 20 percent, and preferably less than several percent. If the error term for the denominator is large, the limits are much more difficult to calculate, and they are not symmetrical around the mean (Finney, 1971).

3. Counting efficiency: Optimally 50 percent for 2π geometry, but generally about 20 percent for alpha spectroscopy.

4. Background counting rate: The background error and sample counting error are propagated by the square root of the sum of the squares. The background for alpha spectroscopy is generally low (counts per hour or less) and stable enough that backgrounds and/or blanks are only run about once a week or less. Thus, there is the potential for actual errors in the blank count that is used to correct the sample gross count to a net count, if the chamber is contaminated.

5. The counting time for both the sample and background or blank impacts the sensitivity as a result of the counting error calculation. The counting error or standard deviation is generally assumed to fit the normal distribution with the variance equal to the total counts (i.e., standard deviation equal to the square root of the total counts). Thus, doubling the counting time reduces the percentage counting error by the square root of 2 (100 counts ± 10, versus 200 counts $\pm 10\sqrt{2}$). Counting times for low-level alpha analyses are normally 1000 min. (Johns, 1975).

Most calculations of counting error and thus statistics (e.g., Johns, 1975) assume the applicability of the normal distribution. Nuclear disintegration or counting statistics are basically described by the binominal distribution (Evans, 1955, and Jarett, 1946). It is only through generalizations and assumptions that the normal distribution is applicable. The basic assumption of concern for low level determinations of long half-life radionuclides is the accumulation of sufficient counts for the transition from the Poisson to the normal distribution. The minimum value normally stipulated is 20 counts, below which the Poisson is too skewed to be approximated by the normal (Evans and Jarett). Jaffery (1960) stipulates a value of 100 counts.

For a mean of 20 counts, the mode of the Poisson is 19 versus the mean and mode of 20 for the normal (Jarett, 1946). Figure 3 is a cumulative frequency plot for a mean of 10 events. The cumulative 50 percent point for the Poisson is about 9, versus 10 for the normal distribution.

Most of the minimum detectable activities (MDA)* are associated with net sample counts of about 10 above a background count of 0 to 5 - where both counting times are about 1000 minutes.

Equation 1 indicates the calculation for the probability, P, of x events occurring for the Poisson distribution, where m is the true value

$$P(x) = \frac{m^{x} \exp{-m}^{**}}{x!} \qquad (1)$$

Equation 2, using the same nomenclature, indicates the probability P (x) for the normal distribution

$$P\ (x) = \frac{1\ \exp-(x-m)^2/2m}{(2\ \pi m)\ 0.5} \qquad (2)$$

For an assumed mean or true value of m = 2, the probability of occurrence of a value of two is similar for the two distributions (i.e., 27.4% for the normal, versus 27.1% for the Poisson). But, the probability values of one or three occurring differs by about 20 percent for the two distributions (the values for the normal distribution are integrated between x plus and minus one-half).

It is difficult to assess the full impact of the limitation of the assumptions in using a normal distribution. But for samples near the MDA, if results related to counts of 10 or less events are used, it appears that the errors in the counting error statements and in actual results could be several tens of percent.

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* For these purposes MDA is used as a general term to indicate the defined detection limit. No attempt is made to distinguish between the original sample, a prepared sample, or curies versus counts.

** $\exp{-m} = e^{-m}$.

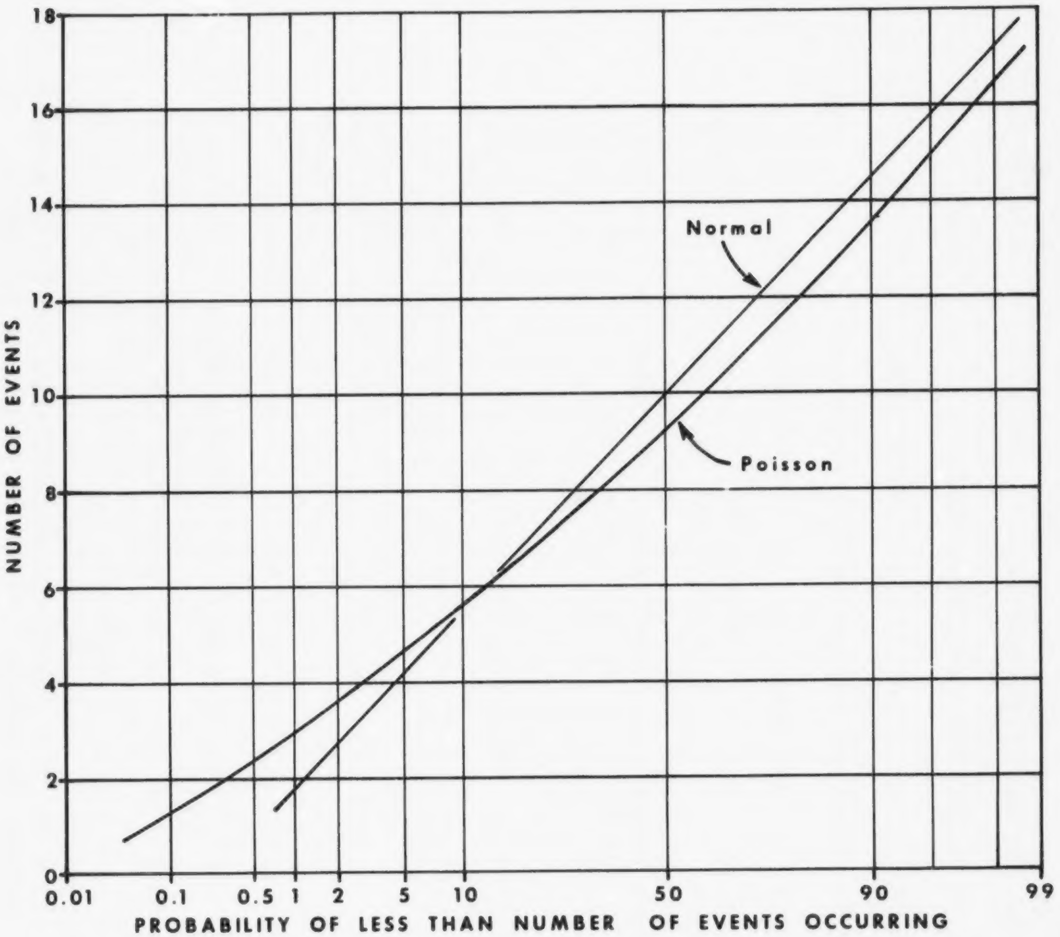

Figure 3. Cumulative frequency plot for a true value of 10.

Johns (1975) presents equations for calculating the plutonium-239 activity in a sample and the associated counting error. The calculations use the ratio of the plutonium-236 tracer added to the sample to the recovered plutonium-236, rather than an actual detector efficiency.

The counting error equation is a propagation of error of the square root of the sum of the squares of the coefficients of variation for the following parameters:

- Sample count for plutonium-239
- Reagent blank count for plutonium-239
- Sample count of plutonium-236 tracer
- Reagent blank count for plutonium-236 tracer

This results in a rather complete analysis of the counting error for a sample. For samples near the MDA, the sample count is probably around 10, and thus has the previously noted limitations of not being normally distributed. The same is true for the background or blank count for all analyses. Thus, the error term may not be truly representative by up to tens of percent for the two values.

In talking to personnel from various laboratories, it was discovered that some do not subtract an instrument background and few subtract a reagent blank background. The significance of errors associated with these practices depends on the level of sample activity, as well as the degree of possible contamination of the counting instruments, reagents, laboratory glassware, and tracer solution. Given the potential for errors, it is prudent not only to subtract background, but also to run reagent blanks containing the tracer, and use this blank as the background.

A general review of the effects of various actual numbers would indicate a potential for a misrepresentation of the error term by up to 30 percent for values near the MDA (assumed to be about 10 counts). The calculations indicate a nominal MDA of about 20 fCi/sample; assuming 1000-min count time, high chemical yield (about 90 percent), and low background (0-5 counts in 1000 min). Consideration of the assumptions indicates that the actual MDA varies from sample to sample, if it is based on counting error. Thus, single values are only nominal estimates based on representative results.

Sill (unpublished document) presents a counting error evaluation which includes several additional factors. Although Sill included error estimates for the tracer standardization and for correcting the tracer for decay since standardization, these errors are a small part of the total error. The error associated with the sample count accounts for well over 90 percent of the total error for his estimate. Furthermore, the potential error of assuming the background is zero overshadows the contributions from these other errors.

MDA's from several organizations are summarized in Table 16. Many authors do not report MDA's, and many that do, report them in terms of their specific sampling and analysis parameters (e.g., pCi/g or nCi/m² for soil, or pCi/m³ for air). The basic MDA for plutonium analysis by alpha spectroscopy is based on the amount of plutonium present on the electroplated sample and the count time. Inclusion of the chemical yield and sample size results in secondary MDA's.

Another approach for evaluating the MDA is to consider results of samples that contain essentially no plutonium. Table 17 from Krey and Hardy (1970) presents results from two samples, one collected prior to 1945 and the second collected from a depth of 90 cm in 1970. The analyses were performed on 100-gram samples and the counting errors are only one standard deviation. Thus, the minimum numbers 0.0001 to 0.0003 dpm per gram relate to 10 to 30 fCi per sample (e.g., 0.0001 dpm/g x 10^3 fCi/2.22 dpm x 100 g/sample x 2 sigma = 10 fCi per sample).

The results for the Woodcliff Lake sample are surprisingly high. Krey and Hardy note the probable cause as contamination, either during collection or analysis. Two of the TLW values are noted as suspect.

TABLE 17. PLUTONIUM IN BLANK AND LOW-LEVEL SAMPLES
(From Krey and Hardy, 1970)

Sample	Laboratory	dpm per gram Plutonium-239	Plutonium-238
Pre-bomb (Collected before 1945)	HASL	0.0003 ± 100%	0.0002 ± 100%
"	IPA	0.0001 ± 100%	0.0001 ± 100%
"	IPA	0.0001 ± 100%	0.0001 ± 100%
"	TLW*	0.0196 ± 7%	0.0054 ± 14%
"	TLW	0.0001 ± 100%	0.0001 ± 100%
Woodcliff Lake, N.J. (Collected below 90 cm	IPA	0.0046 ± 7%	0.0001 ± 100%
in March 1970)	IPA	0.0043 ± 6%	0.0001 ± 100%
"	TLW	0.0071 ± 9%	0.0009 ± 53%
"	TLW*	0.0468 ± 5%	0.0001 ± 100%
"	TLW	0.0055 ± 25%	0.0002 ± 100%

* Suspect value

TABLE 16. SUMMARY OF MDA'S FOR PLUTONIUM IN ENVIRONMENTAL SAMPLES

Investigator	Definition of MDA	Basic MDA fCi/sample	Soil fCi/g	Soil Sample Size (g)	Air fCi/m^3	Air Sample Size (m^3)	Water fCi/l	Water Sample Size (l)	Tissue fCi/g ash	Tissue Sample Size (g/ash)
Denham and Waite (1974) Survey[a]	---	--- ^{238}Pu	3	(0.03-500)	10^{-3}	($5x10^{-4}$-0.1)	5	(0.5-50)		
		^{239}Pu	3	(0.4-30)	$5x10^{-3}$	(10^{-3}-0.1)	10	(0.5-50)		
Johns (1975)	$\bar{x}\leq 2\sigma$	20	20 4	1g 10g[b]						
Poet and Martell (1972)		Only report 1 σ error								
AEC (1973a), Sedlet et al. (ANL)	$\bar{x}\pm 2\sigma$	∿5(?)			10^{-4}	25-60x10^3	0.1 0.5	45 10		
Robinson et al. (1975)			100	10						
E. Geiger (Eberline Instrument Corporation Verbal 5/7/75)	$\bar{x}\pm 2\sigma$	20		10						
Krey and Hardy (1970) (estimated by Bernhardt)		∿10-55								
Smith and Black (1975)	$\bar{x}\pm 1\sigma$	20								
McDowell et al. (1973)	Sample=Bkgd[c]	140	(liquid scintillation, alpha spectrometry)							
Majors et al. (1974)	$\bar{x}>1\sigma$(?)	20								
		15,000	(Gamma spectrometry for ^{241}Am)							
McBryde (McClellan, Verbal, 1975)	$\bar{x}>1\sigma$	0.06-6	(Mass spectrometry with sophisticated and routine chemistry)							
Church (1974) REECo et al.		∿300,000	3,000 ^{241}Am in soil	100						

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a. Summary paper of AEC Contractor techniques. Single value is considered typical; numbers in sample size column indicate the range.

b. Present yield on 10-gram samples is only about 50%.

c. Sample countrate equals background countrate.

A possibly unfair conclusion would be that "zero" for these samples ranged from 10 to 55 fCi per sample, assuming the two TLW samples can be excluded, which probably would not be the case for unknown samples.

Robinson et al. (1975) report 50 values for a background sample which should have contained only 0.2 fCi/g of plutonium-238. Their values range from 0 to 765 fCi/g, based on a 10-gram sample (no background subtracted). The standard deviation for a single result is a counting error of 40 fCi/g. Two times the counting error, 80 fCi/g, essentially is equal to the average of the 50 results, 77 fCi/g. Based on their analysis, they picked 100 fCi/g as the minimum reporting value for reliable results. This relates to an MDA of 1000 fCi per sample (100 fCi/g x 10g). The intent of the reported project was to assess plutonium-238 contamination significantly above the background level of 0.2 fCi/g for a 12-in core.

Figure 4 is a histogram of Robinson's et al. data. It can be seen that 20 percent of the values (blank or background) are above the MDA of 100 fCi/g.

The optimum MDA, assuming essentially zero background, 1000-min counts, and ignoring the limitations of the statistical assumptions, is about 10 fCi. Practically, a more reasonable minimum is 20 fCi. The value of 20 fCi relates to about 3×10^{-13} g of plutonium-239 or 1×10^{-15} g of plutonium-238. Malaviya (1975) indicates a theoretical capability for mass spectrometry of 10^{-18} g.

In summary, there are several means of defining the sensitivity of analyses, or minimum detectable activity. The techniques that give the lowest MDA's that are reasonably valid are based on the 2- or 3-sigma counting error. The EMSL-LV technique (Johns, 1965), defines the MDA value as the mean value equal to the two-sigma error. Others sometimes use three times the background counting error, which generally gives results similar to Johns (1975). In most instances when mean sample results are below or equal to the MDA, they are expressed as less than the MDA.

It should be recognized that most less than values are only a 50 percent probability statement. That is, 50 percent of the time the statement is wrong. A reasonable minimum MDA is about 20 fCi per sample; i.e.,the counting error is 100 percent at the 2-sigma or 95 percent confidence level. A more realistic MDA statement , given the limitation of the statistical assumptions, would be less than 20 fCi plus 1- or 2-sigma, i.e., 30 or 40 fCi per sample. These values are in essence per sample planchet, after electroplating. If the chemical (tracer) yield is only 50 percent, the values actually are 40, 60, and 80 fCi per original sample.

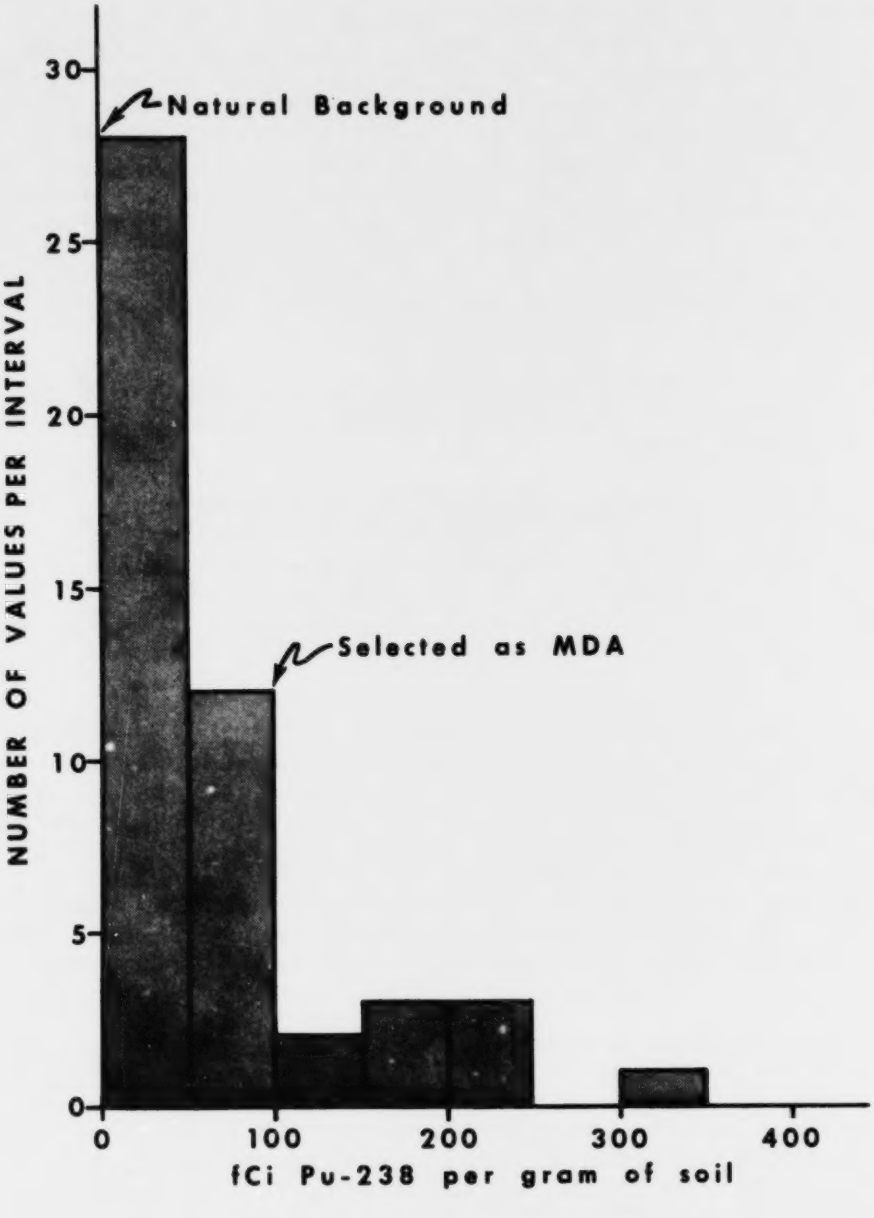

Figure 4. Histogram of blank or background ^{238}Pu soil samples.

Table 18 converts the sample MDA into MDA's for various environmental samples. It is evident from the table that present analytical techniques can detect plutonium at concentrations well below the standards.

SAMPLE TYPES

The chemical and physical characteristics of samples in part determine the dissolution technique for getting the plutonium in solution for analysis, and the steps in the analysis that are necessary to remove elements that interfere with subsequent steps in the analysis, especially electroplating. The various refractory compounds of plutonium, and the generally low solubility of many plutonium compounds, requires emphasis on the complete dissolution of the sample material to assure dissolution of any associated plutonium. If there is residual sample material, there is concern that there may be plutonium in the residual. Sill et al. (1974) and Sill and Hindman (1974) indicate that non-fusion techniques may leave up to 40 percent of refractory plutonium in the undissolved residual.

The refractory nature of plutonium in the sample is related to several factors, including the following:

1. History of the source of the plutonium in the sample and its particle size distribution. For example, Rocky Flats and global fallout plutonium generally are amenable to leaching techniques (Krey and Hardy, 1970), while plutonium from many of the NTS tests appears to be in the form of generally insoluble discrete particles.

2. Sample preparation techniques, such as firing to remove soil organic matter, can produce refractory plutonium (Sill and Hindman, 1974). Sill and Hindman indicate that temperatures of about 700-1000°C produce refractory plutonium.

3. The nature of the sample material, particularly soil samples, can have an impact on the dissolution. Limestone and coral are largely calcium carbonate and can be dissolved rather readily with nitric or hydrochloric acid (AEC, 1973; Wessman et al., 1974; and E. Geiger, Eberline Instruments Corporation, verbal communication, May 6, 1975). The amount of calcium in a limestone or coral soil can produce interferences in a fusion-type technique. Iron oxides also are less prevelent in limestone and coral, resulting in less interference from iron.

4. Most soils are composed of from 50 percent to 80 percent sandstone. Thus, there is a large amount of undissolved residual material from leaching techniques,

TABLE 18. MINIMUM DETECTABLE CONCENTRATION

	SAMPLE VOLUME	UNITS[a]	MINIMUM DETECTABLE CONCENTRATION BASED ON SAMPLE MDA OF		POPULATION (INDIVIDUAL) RPG[a]	RATIO: 20 fCi MDA/RPG
			20 fCi	40 fCi[b]		
AIR SAMPLES						
10 cfm - 1 day	400 m^3	fCi/m^3,x10^{15}uCi/cc	0.050	0.10	60	0.0008
10 cfm - 3 day	1,200 m^3	fCi/m^3,x10^{15}uCi/cc	0.017	0.03	60	0.0003
10 cfm - 7 day	2,800 m^3	fCi/m^3,x10^{15}uCi/cc	0.007	0.014	60	0.00012
40 cfm - 1 day	1,600 m^3	fCi/m^3,x10^{15}uCi/cc	0.013	0.025	60	0.00021
40 cfm - 3 day	4,900 m^3	fCi/m^3,x10^{15}uCi/cc	0.004	0.008	60	0.00007
WATER SAMPLES						
1-liter	1-liter	fCi/lor10^{12}uCi/ml	20	40	5×10^6	4×10^{-6}
5-liter	5-liter	fCi/lor10^{12}uCi/ml	4	8	5×10^6	0.8×10^{-6}
SOIL						
1-gram	1 gram	fCi/g	20	40	------C	
10-gram	10 gram		2	4	------	
100-gram	100 gram		0.2	0.40	------	

a. 1fCi/m^3 =10^{-15} uCi/cc. These values are n fCi/m^3 or n x 10^{-15} uCi/cc.

b. An MDA of 40 fCi is the same as the MDA for 20 fCi with a 50% chemical yield. A yield of about 100% has been assumed. The MDA's are for 20 or 40 fCi per original sample quantity.

c. There are no Federal Standards for Pu in soil. Colorado stipulates 2 dpm or about 1 pCi/g. The RPG's are those from 10CFR20, for the most limiting form.

with a potential for retained plutonium. Furthermore, there is the potential for plutonium oxide to occur in a siliceous matrix.

5. Liver and kidney samples present analysis problems analogous to or worse than soils because of the presence of heavy metals, other than plutonium. Due to various interference mechanisms, chemical yields at times are close to zero (J. Mullins, EMSL-LV, verbal communication).

6. In essence, analysis of air samples presents the same difficulties as the analysis of soil samples. The plutonium on air filters is associated with essentially the same material, with possibly a smaller particle size distribution, as the plutonium in soil (e.g., 0.05 pCi/g x 100 $\mu g/m^3$ = 0.005 fCi/m^3, roughly one-tenth of ambient air background). The air sample filtering material may present additional analytical difficulties (e.g., fiberglass air filters are difficult to dissolve and have metals that interfere with the analysis of plutonium).

REVIEW OF ANALYTICAL TECHNIQUES

As in most areas of life, there are few absolute generalizations that can be applied to plutonium analytical techniques. Recognizing this, but also recognizing a need for categorization, plutonium analytical techniques may be divided into three basic techniques for getting the plutonium in solution and four techniques for plutonium quantification.

The techniques for placing the plutonium in solution are:

1. Leaching: The technique generally is related to that of (or represented by) Chu (1971). The basic technique is to leach plutonium from the sample with HNO_3 and HCl. The sample generally is digested for several hours at boiling temperatures. The technique has the advantage of being able to treat large soil samples, nominally 100 g, but up to 1000 g or more. Also, the technique is less likely than other techniques to dissolve interfering elements along with the plutonium. A significant volume of residue remains after the leaching. The technique can be conducted by normal radiochemistry technicians. The disadvantage is the potential for not having dissolved all the plutonium, or having it in an available chemical state.

2. Acid dissolution: This technique can be considered an advanced acid leach. The basic difference is the use of additional HF (the leach technique may use some HF) and the increased digestion and treatment to the point

where essentially the whole sample is placed in solution, with only minimal residue. Hydrofluoric acid has the ability to dissolve silica, and also increases the solubility of refractory oxides, forming fluoride complexes (Sill et al., 1974). The technique generally is amenable to sample sizes of 10 to 15 grams of soil or ash, although the implementation of the technique is easier with 1-g samples. Mullins (EMSL-LV, verbal communication) notes that the treatment of 10-g samples requires the use of professional personnel, or increased supervision of technicians. The increased dissolution of the sample results in increased dissolution of interfering metals. There is an increased probability of dissolving refractory plutonium, but there is still some uncertainty about complete dissolution and chemical availability of the plutonium (Mullins, verbal communication, January, 1975, and Sill and Hindman, 1974). The EMSL-LV method generally is representative of this technique (Johns, 1975).

3. Fusion: Sill (1969) and Sill and Williams (1969) have developed the basic technique of a pyrosulfate fusion for placing uranium and the transuranium elements in solution. The tentative EPA Reference Technique is essentially identical to this method, Hahn et al. (in press). Furthermore, this technique is used to check the efficiency of other techniques (Sill et al., 1974). This method generally is limited to 10-to 15-g samples because of available equipment size limitations. Furthermore, the method requires a high degree of technician proficiency, generally professionals.

Sill et al. (1974) present a summary of the concept of several analytical techniques. The following discussion is based on their review.

Basically, analysis can be broken down into the following phases:

1. Sample dissolution and addition of tracer.
2. Chemical separations to isolate desired elements from interfering elements through precipitation, volitalization, and ion-exchange.
3. Electroplate (or by other means) place sample on planchet or metallic disk (or place in solution for liquid scintillation).
4. Count sample by appropriate technique, such as alpha pulse height analysis.

5. Calculate sample activity and estimate analytical error term, based on tracer yield and blank or background count rate.

Church et al. (1974), Majors et al. (1974), and Gilbert and Eberhardt (1974) report data on americium-241 analysis by gamma spectrum analysis. Values for plutonium-239 can be estimated from assumed or calculated plutonium-239/plutonium-241 ratios, based on radiochemistry. Gamma spectrum analysis for americium-241 essentially requires no sample preparation or radiochemistry. The sample is dried and placed in a standard container. Given the relatively low gamma energy (60 keV) and photon abundance, the sensitivity of the method is not adequate for ambient concentrations. The sensitivity is about 100,000 fCi per sample, plus or minus about a factor of five depending on the other gamma emitters present and the counting time. The technique is amenable to samples of roughly 100 grams (e.g., sensitivity roughly 1000 fCi/g or 1 pCi/g). Piltingsrud and Stencel (1973) present similar information for phoswich detectors.

Each of the five analytical steps are discussed in detail below.

Sample Preparation and Dissolution

Sample preparation usually consists of drying the sample at about 100 to 120°C. This normally is the weight basis for reporting results. The difference in weight between air dried (Krey and Hardy, 1970) and oven dried weights may range up to 15 percent (Bliss, EMSL-LV, verbal communication). If there is a significant amount of organic material and roots, the sample is then heated in a muffle furnace to 400°C (Sill et al., 1974) or to 600°C or more; or the material may be burned off with a blow torch, (Bishop et al., 1971). Sill et al. (1974) and Sill and Hindman (1974) express the concern that the high temperatures will increase the refractory nature of the plutonium. This can affect dissolution for silica soils, but apparently does not for coral-type soils.

Many authors recommend sieving the samples subsequent to ball milling them (e.g., Sill et al., 1974 and Krey and Hardy, 1970). Gilbert (verbal presentation at May 1975 NAEG meeting) noted a disparity between sieved and non-sieved aliquots of groups of samples. Gilbert's comments were not conclusive, but indicated a concern for sieving. Possibly a disportionate amount of fines containing plutonium are electrostatically bound to the larger particles.

The following discussion of sample and plutonium dissolution primarily relates to soil samples. However, subsequent to dissolution or combustion of the filter, it also can be related to samples of airborne particulates. The dissolution of ashes from various biological samples is similar.

Most of the techniques are based on dissolving the sample in concentrated hydrochloric and nitric acids. The digesting times and temperatures vary. One of the most significant variations between techniques is the amount and concentration of hydrofloric acid used. Hydrofloric acid is recognized for its ability to dissolve silica, the predominant material in most soils.

Hydrofluoric acid is used to dissolve and break down the silica and soil matrix. The "dissolution" versus leaching processes call for an excess of HF. The leaching processes use little, if any, HF. The HF also acts as a catalyst for breaking down the plutonium and getting it into ionic form in solution.

Sill et al. (1974) and Sill and Hindman (1974) stress the difficulty and necessity of getting the plutonium into a monomeric, ionic form. Plutonium is prone to forming colloids and complex ions. Thus, dissolution alone is not sufficient--it must be in ionic form. Mullins (EMSL-LV, verbal communication) notes that sometimes the miscellaneous heavy metals in liver or kidney tissue can form complexes with the plutonium tracer, resulting in a zero tracer yield. Emphasis must be placed on insuring that the sample and tracer are in chemical equilibrium; e.g., the same ionic state.

It is important that the tracer be added at the right time. If tracer is added to an empty beaker, it may bind to the beaker. The resulting low yield does not reflect the recovery of sample plutonium. Also, if tracer is added too late in the process, the yield will not reflect plutonium losses prior to the tracer addition. In any case, there is always uncertainty as to whether the tracer truly interacts with the plutonium in the sample. The probability is that the tracer may exhibit a yield higher than that of the sample plutonium. But it is possible that the tracer plutonium may also be lost while the sample plutonium is still tied to the sample, thus indicating a yield lower than that achieved for the sample plutonium.

Sill et al. (1974) recommend a combination potassium fluoride and pyrosulfate fusion subsequent to the previously indicated acid treatment to ensure the complete dissolution of the sample and associated plutonium. They note that sodium carbonate or hydroxide fusions do not guarantee complete dissolution, and that the necessary subsequent steps often result in yields of less than 50 percent to as low as 2 percent.

The anhydrous potassium fluoride fusion (in a platinum dish) is used to insure the complete dissolution of siliceous material. The pyrosulfate fusion is used to insure complete dissolution of nonsiliceous materials, especially high fixed oxides (plutonium) along with the volatilization of hydrogen fluoride and silicon tetrafluoride. Except for a small amount of barium sulfate, the pyrosulfate cake resulting from the fusions can be readily dissolved in dilute HCl.

Sill et al. (1974) recommend that the fusion technique be used to check undissolved residuals from other dissolution techniques, resins, and various laboratory equipment. They emphasize the validity of checking residuals versus relying on tracer yields or duplicate analysis by the fusion technique.

Chemical Separations

Sill et al. (1974) precipitate the alpha emitters (radium through californium) with barium sulfate. The various elements are extracted from the solution through control of valence states and solvent extraction. There are several steps where care must be taken to prevent the hydrolytic precipitation of plutonium, the carry-over of iron or quadrivalent cerium with plutonium, and subsequent electrodeposition interference.

Sill et al. (1974) note that for soil samples, calcium is the worst source of interference for the barium sulfate precipitation, because of its relatively high concentration (∿3%) in most soils. If the calcium present in 10 grams of soil precipitates as calcium sulfate and is filtered off, it probably will carry most of the alpha-emitter ions with it. The acidity of solution can be increased by the addition of HCl, but this affects the barium sulfate precipitation. Apparently, these losses are acceptable for plutonium up to a value of about 5 percent calcium in a 10-gram soil sample. A dissolution of the initial barium sulfate precipitate with reprecipitation is necessary to remove small quantities of calcium and other ions which would interfere with electrodeposition and alpha resolution from the deposited sample.

Sill et al. (1974) note various modifications for recovery of the alpha emitters other than plutonium. The basic method is oriented to plutonium.

Sill et al. (1974) report the activity associated with sample residuals and the various pieces of analytical hardware. This data can be used to assess sources of cross-contamination and critical points where sample activity may be lost.

Talvitie (1971) describes the basic method for ion exchange separation of the elements. The technique emphasizes the separation of iron to prevent interference during electrodeposition of plutonium. Talvitie's method is used by Johns (1975).

Bentley et al. (1971) describe the LASL solvent extraction technique. The plutonium is extracted into di-2-ethylhexyl orthophosphoric acid (MDEPH).

Electrodeposition

Electrodeposition generally is used to produce the uniform, essentially weightless, deposition needed for alpha spectroscopy.

The sample must be essentially infinitely thin to minimize self-absorption, or energy degradation, of the alpha particles. Evaporation of solutions on a hot plate does not produce an adequately uniform deposit (Talvitie, 1972). Several alternative techniques include liquid scintillation counting as described by McDowell et al. (1973), and co-precipitation of plutonium with trace amounts of lanthanum carrier (Lieberman and Moghissi, 1968, and Butler et al., 1971). Mass spectrometry also eliminates the need for electrodeposition, but equipment and personnel requirements generally are beyond the resources of most laboratories. Most laboratories utilize alpha spectroscopy after electrodeposition of the sample. Although electrodeposition entails inherent problems, it generally results in a higher quality alpha spectrum than liquid scintillation or plutonium co-precipitation. These alternate techniques, along with mass spectroscopy, will be discussed at the end of this section.

There are several basic potential problems associated with electrodeposition. One basic problem in all radiochemistry procedures is residual contamination of equipment from prior sample analysis. This is especially true with electrodeposition equipment. Talvitie (1972) describes a technique based on disposable electrodeposition cells to minimize this problem. This process is used by EPA/EMSL (Johns, 1975).

Changes in the electrolyte pH during electrodeposition and various elements, such as iron, interfere with electrodeposition, increase the thickness of the deposit, and result in low and variable yields. Talvitie (1972) describes recovery from 1M ammonium sulfate at pH 2 in a period of about 40 minutes. It is recommended that the iron content be less than 0.1 mg.

Puphal and Olsen (1972) describe recovery from ammonium chloride-ammonium oxalate electrolyte over about 50 minutes. They discuss the use of a chelating agent to reduce the interference of some cations, and fluoride to alleviate the interference from iron, aluminum, thorium, and zirconium. But they noted that the presence of even microgram quantities of rare earths can cause serious interference if fluoride is added.

Sill (verbal, EPA/NERC-LV Workshop, now EMSL-LV, April 3, 1974) notes that using methyl red as a pH indicator prior to electroplating results in the possible formation of plutonium hydroxide. Although the pH is corrected prior to electroplating, the plutonium hydroxide may not dissociate and go back into solution. Thus, Sill recommended using thymol blue as the indicator. Hahn et al. (in press), Johns (1975), AEC (1974), and Sill and Hindman (1974) use thymol blue.

Sill et al. (1974) note that subsequent to reasonable dissolution, electrodeposition is by far the step with the greatest potential for loss of the sample. Electrodeposition of electropositive elements such as the actinides depends on deposition of hydroxides by hydroxyl ions produced electrolytically at the cathode. All metal ions forming insoluble hydroxides may be expected to electrodeposit to some extent, degrading the sample plate and thus the alpha spectra. Furthermore, if precipitates are formed during pH adjustments, the element being determined may coprecipitate more effectively than if it were alone and therefore not be available for electrodeposition. This is especially worrisome with high pH's around 4.8 to 6 (i.e., methyl red) which is the justification for recommending the use of thymol blue (pH 1.2 to 2.8). Sill et al. (1974) recommend using the salmon-pink end point of thymol blue (pH 2.0).

Sample Counting Techniques

There are four basic counting techniques: Alpha counting of solid samples, liquid scintillation counting of alpha particles, gamma counting for americium-241 (estimate of plutonium-239), and mass spectrometry.

Various types of alpha counters can be used for gross alpha counting. Lieberman and Moghissi (1968) propose a plutonium method with separations appropriate for gross alpha counting. But as in all gross counting techniques, there is the potential for error as a result of inadequate separation. Bains (1963) notes that ambient-level samples, purified to the extent of about one net count per hour, often contain sufficient natural activity to affect low level results. Bains concludes that spectrometry is needed for low-level alpha work.

Sill and Hindman (1974) and Hahn et al. (in press) suggest standardizing tracer in 2-π alpha counters prior to standardizing alpha spectrometers. The need for cross checking electrodeposition samples (standardization), due to the uncertainties, with standards made up from solutions evaporated on counting disks is stressed. Alpha spectrometers are only calibrated as a general cross check, because normally sample activity estimates are derived from the observed tracer counts versus the amount added (i.e., yield and counter efficiency are considered in a single parameter).

Due to the degradation of alpha particles in the electroplated source, and the separation distance between sample and counter, alpha spectrometers normally operate at 20-30 percent efficiency (Mullins, verbal, February 1975, and Sill and Olson, 1970). Sill and Olson (1970) and experience at EMSL-LV (Mullins, verbal, February 1975) stress the need to consider potential detector contamination from alpha-active daughter products of the sample activity. The concern for contamination relates to the alpha recoil of nuclides, possibly in connection with the volatility of nuclides. Polonium-210 appears to present the greatest hazard. Preheating the plate prior to counting appears to minimize the problem.

Alpha spectroscopy is only appropriate for analyzing the plutonium isotopes of mass 236, 238, 239, 240, and 242. The isotopes plutonium-236 and -242 normally are not found in the environment in significant quantities, and are used as tracers. The alpha energies of plutonium-239 and 240 are so close together they cannot be distinguished by alpha spectroscopy. Plutonium-241 is a beta emitter, and thus, although it is the plutonium isotope normally present in the environment in the largest curie quantities, it cannot be determined by the normal plutonium quantitation techniques. Plutonium-241 quantities normally are estimated from assumed isotopic ratios, from the estimated ingrowth of its progeny americium-241, or by mass spectroscopy.

McDowell et al. (1973) describe a liquid scintillation method for low-level alpha counting. The method has the advantage that electrodeposition, with the associated problems of various interferences, is excluded. But due to the inherently higher background of the liquid scintillation counter, its normal sensitivity is higher than that for solid state alpha spectroscopy. The increased background of liquid scintillation is partially offset by the increased counting efficiency.

McDowell et al. (1973) indicate an alpha counting efficiency of 100 percent with energy determination capability of ±0.1 MeV. The MDA for Pyrex sample tubes is reported as 1 dpm (0.5 pci-whereas for quartz sample tubes the level is reduced to 0.3 dpm (0.14 pCi). If pulse shape discrimination is used, a value of 0.02 dpm or 10 fCi appears attainable.

The normal system background is reported as 1 cpm. This can be reduced to 0.3 cpm by using quartz sample tubes. Pulse-shape discrimination, which requires sample deoxygenation can reduce the background to 0.01-0.05 cpm.

It appears that the sensitivity or MDA has been set equal to the background. Assuming the sample and background counting times are equal, this is equivalent to an MDA where the two-sigma error is equal to or less than the MDA.

Energy discrimination or resolution is such that plutonium-236 tracer and plutonium-239 can be counted simultaneously. McDowell et al. (1973) indicate that background can be determined simultaneously from adjacent channels (away from actual channels of interest). Although this alleviates the need of separate background determinations, it has the potential error of overlooking separation errors, and background due to reagent or equipment contamination. It also is noted that if uranium is not separated from plutonium, impurities in the scintillator may cause overlap of the uranium-234 and plutonium-239 peaks. The uranium-238 peak can be used to estimate the uranium-234 interference.

McDowell et al. (1973) note that iron and other metals do not interfere with liquid scintillation counting, since electrodeposition, the point of interference, is not necessary. Thus, simpler, less time-consuming separation steps may be used (1 hour versus 10 or more hours), and the uncertainties and potential yield reduction associated with electroplating is eliminated.

McDowell et al. (1973) indicate that even with uranium in 10^8-fold mole excess over plutonium, quantitative separation and recovery can be obtained. In summary, it appears that liquid scintillation counting can be used to quantitate plutonium at ambient concentrations, but the equipment and techniques are more sophisticated than normally available at most laboratories.

Lieberman and Moghissi (1968) and Butler et al. (1971) describe a technique using trace amounts of lanthanum to coprecipitate plutonium. The essentially weightless precipitate is collected on a membrane filter and is amenable to alpha spectroscopy analysis. There is some degradation of the alpha spectrum, but apparently most samples can be quantitated easily. If there is too much mass in the precipitate, it can be dissolved and purified. Robinson et al. (1975) report cross-check results between the EPA laboratory in Montgomery, Alabama* which uses this technique and Mound Laboratory. The results show good reproducibility. The co-precipitation technique apparently has received only limited use, but appears to have definite utility, either for those who do not have electrodeposition capability or who would prefer an alternate technique.

Mass spectrometry (MS) provides isotopic data not available from alpha spectroscopy (AS) (plutonium-240 and -241) and it also has greater potential sensitivity than AS. In essence, it is based on counting the number of atoms of a given mass. Thus, its sensitivity, if converted to pCi/g, is greater for long half-life nuclides than for short half-life nuclides, because more mass is present for a given curie quantity. Mass spectroscopy often is used only to determine isotopic ratios, but if a tracer is used, it can be used to quantitate results. It often is used to supplement alpha spectrometry results.

For the long half-life isotopes of plutonium, MS has the potential for several orders of magnitude sensitivity greater than AS. For the present day optimum state of the art, as practiced by the McClellan Air Force laboratory, the routine sensitivity of MS is about an order of magnitude greater than AS sensitivity.

Mass spectroscopy is based on determing the number of atoms of a given mass number. Thus, just as in AS, chemical separations are necessary to remove interfering elements. These interfering elements may be either elements with isotopes of the mass of interest (e.g., uranium-238 and plutonium-238) or isotopes that can be combined with the MS filaments to provide interfering mass units. Thus, more sophisticated chemistry (not justified for routine samples) can be used to increase the sensitivity by roughly an order of magnitude.

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*Eastern Environmental Radiation Facility (EERF).

The sensitivity of MS for plutonium-238 is less than that for AS. This is because of the short half-life of plutonium-238 and interference from traces of uranium-238. Also, because of the short half-life of plutonium-236, plutonium-242 is the preferred tracer. The present state-of-the-art routine sensitivity for MS is about 10^{-13} g of plutonium (Merrell, Air Force McClellan Central Lab, verbal communication May 1975). This can be reduced to about 10^{-15} g with special chemistry techniques. The value of 10^{-13} g is equivalent to 6 fCi of plutonium-239.

Isotopic ratios determined by MS often can be used to determine the source of environmental contamination. Evaluations by Krey (1976) and Krey et al. (1975) illustrate the utility of isotopic ratios, in conjunction with quantitative results, to distinguish the source of contamination.

Americium-241 can be quantitated by either gamma counting with NaI(Tl) wafers (Majors et al.,1974) or Ge(Li) semiconductor detectors. Quantitation is based on the 60-keV photon. Plutonium-239 may then be estimated based on the plutonium-239/americium-241 ratio determined from radiochemistry analysis of a selected number of samples. The sensitivity of the americium-241 method is dependent on the associated gamma emitters in the sample and the counting time. Brady (REECO, verbal presentation, NAEG, May 1975) indicated a plutonium-239 sensitivity of about 50 pCi/g based on a plutonium-239/americium-241 ratio of ten. This relates to an americium-241 sensitivity of about 500 pCi/g. Brady noted the plutonium/americium numbers agreed within about 50 percent with chemistry numbers. The complications of plutonium-239:241 ratios and americium-241 ingrowth time have to be considered.

Piltingsrud and Stencel (1973) report on an X-ray measurement technique for the low-energy X-rays from plutonium-239 and americium-241. The detector is based on a sandwich of a NaI(Tl) crystal backed by a CsI crystal. The two detectors have different pulse rise times, thus photons interacting with both detectors can be discriminated from low energy photons (X-rays) interacting with the NaI detector. The sensitivity is about 20 pCi/g for 500 g samples - 10,000 pCi per sample (plutonium-239 + americium-241).

The detector does not distinguish between plutonium-239 and americium-241 or other low energy X-ray emitters. However, except for plutonium-238 contamination (normally lower than plutonium-239), most of the X-rays would be from plutonium-239 and americium-241.

Calculation of Sample Activity and Estimation of Analytical Error

The calculations are based on multiplying the measured sample activity by the ratio of the known amount of added tracer to the recovered tracer. The counting errors generally are based on propagation of the normal error based on the observed sample and background counts (Johns, 1975).

There are several potential sources of error (not statistical) associated with the various techniques. These include:

1. Use of plutonium-236 tracer, which has a relatively short (2.85-year) half-life. Thus, the standard tracer solution should be corrected for decay subsequent to calibration, and recalibrated periodically.

   In past years plutonium-236 contained a small amount of plutonium-238 contamination. Thus, any initial calibration error would be compounded with time due to the relative increase of the plutonium-238 fraction, due to plutonium-236 decay. The plutonium-238 contamination must be subtracted from plutonium-238 results for samples traced with plutonium-236. Furthermore, the plutonium-236 tracer solution should be purified periodically to prevent interference from the progeny--uranium-232, thorium-228, and subsequent progeny (Sill, 1974).

2. Americium generally is separated from plutonium prior to analysis. The separation factor generally is several orders of magnitude, so although the americium-241 alpha is similar in energy to the plutonium-238 alpha, there should be little problem. But due to the presence of plutonium-241, americium-241 ingrowth must be considered. Mullins (EMSL-LV, February 1975) notes that samples generally are counted within one month of separation.

   Table 19 indicates various plutonium-238:americium-241 ratios. It is evident that americium-241 ingrowth cannot be ignored completely for normal plutonium isotopic ratios.

3. Enough tracer should be added to produce a small counting error for yield estimates. But there is some uncertainty in the necessary amount due to varying yields. Furthermore, if there is too much plutonium-236, its peak can interfere with the plutonium-239 alpha peak. This can be compensated for by using blank reagent samples, including tracer, for background determinations. Sill (1974) and Johns (1975) suggest about 10 dpm per 10-gram soil sample.

TABLE 19. AMERICIUM-241 INGROWTH INTO PLUTONIUM SAMPLES

Activity Ratio[a] Pu-241:Pu-238	Time After Separation (days)	Ci Am-241[b] per Gram Pu-241	Activity Ratio Pu-238:Am-241
166	0	0	0
166	30	0.013	51
166	60	0.038	17
166	90	0.065	10
166	120	0.15	4

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[a] Assume plutonium-241 is 0.5% by weight or 89% by activity of environmental plutonium at the time of release. Assume the weight percent has decayed to 0.25% (about 15 years). Assume the plutonium-239: plutonium-238 activity ratio is 35. Thus, per gram of plutonium, there is 110.3 Ci/g x 0.25% = 0.276 Ci of plutonium-241 and 0.0614 Ci Pu-239/g x 95% x 1 Pu-238/35 Pu-239 = 1.67 x 10^{-3} Ci of plutonium-238. (Putzier, 1966; Krey and Hardy, 1970, and Del Prizzo et al., 1970).

[b] Putzier, 1966, Figure 13.

4. The separation of polonium from samples should be considered. The alpha from polonium-208 tracer and polonium-210 may interfere with plutonium-239.

5. The background measurement technique and time interim between background measurements can be a source of error. The background from reagents, glassware, and the tracer should be assessed. The potential for contaminating counters (especially from polonium) in part indicates how often backgrounds should be taken.

   Grouping together of samples of similar activity levels for analysis minimizes the potentials for errors due to contamination.

DISCUSSION AND COMPARISON OF TECHNIQUES

Table 20 summarizes the dissolution techniques used by various organizations and investigators. As indicated in the previous sections, assuming a representative sample is taken for

TABLE 20. SUMMARY OF DISSOLUTION TECHNIQUES

Reference	Sample Type	Dissolution Method			Comment
		Leach	Acid Diss.	Fusion	
Church et al. (1974)	Soil		x		REECO, NTS
Johns(1975)	Soil		x		ESML
Krey & Hardy(1970)	Soil	x			Rocky Flats et al.
Major et al.(1974)	Vegetation		x		NTS checked residual by fusion
Talvitie(1971)	Soil, Air Filters		x		
Silland Hindman(1974)	Soil			x	KF & pyrosulfate fusion
Sill et al.(1974)	Soil			x	KF & pyrosulfate fusion
Hahn (in press)				x	Tentative Reference Method
AEC(1974)	Soil		x		
Toribara et al.(1963)	Biological Samples	x		x	Carbonate & bisulphate fusion
Markussen(1970)	Environmental Samples			x	K pyrosulphate, Thule samples
Chu(1971) HASL tech.	Soil	x			Proposes using HF and HNO_3 on siliceous material
Essington (1973) LASL	Soil		x		Modified HASL/LASL
Bokowski(1971)	Soil		x		Rocky Flats, Dow Chem. Repeats $HF-HNO_3$ step 5 times
Bains(1963)	Biological Samples		x		Leach plus HF.
AEC(1973)	Coralline Soils	x			Enewetak
Keough and Powers(1970)	Biological Samples		x		
Bentley et al.(1971)	Soil		x		LASL
Lieberman &Moghissi(1968)	Environmental Samples		x	x	Alkali fusion
Corley et al.(1971)	Soil	x			Hanford
Butler et al.(1971)	Soil			x	Study of fusion techniques (after Chu, 1971)
Wessman et al.(1971)	Soil	x	x		
Bishop et al.(1971)	Soil	x		x	May use 1 ml of HF in leach

analysis, the primary analytical concern is dissolution of the sample. Other prime concerns relate to consideration and removal, if possible, of interfering elements and ions (calcium, iron, etc.). This interference is of less concern than dissolution, because generally it is reflected by low tracer yields and is thus accounted for.

Analysis of samples by liquid scintillation counting or mass spectrometry eliminates the need for electrodeposition, the analysis step that has the greatest potential for interference and loss of plutonium (Sill and Hindman, 1974). Liquid scintillation tends to be less sensitive, without using special techniques, than solid-state alpha counting. Thus, although it has potential and possibly should be investigated, it will not receive further consideration in this section.

Mass spectrometry has several virtues for plutonium analysis, including providing information on isotopic ratios and improved sensitivity. But due to its limited use for quantitation of environmental plutonium samples, the high sensitivity for plutonium-238, and the expense for organizations to initiate this type of analysis, it will not receive further consideration. Furthermore, it entails the same concerns about dissolution of the initial sample as alpha spectrometry. Also, it has its own unique problems of separation and removal of interfering substances (iron, uranium, and hydrocarbons).

Sample Size

Sample size considerations, based on analytical sensitivity and the representativeness of the sample have been discussed previously. The analytical sensitivity generally is inversely proportional to the sample size (e.g., sensitivity in pCi/g is 10 times higher for a 1-gram sample than for a 10-gram sample). However, the increased amounts of interfering substances in large samples may decrease the chemical yield and thus the relative benefit of large samples. The discrete or heterogeneous nature of plutonium in samples limits the minimum size for analysis aliquots, depending on the acceptable variability of sample results. This potential variability has been discussed previously, but the actual situation will depend on specific sources of contamination and samples.

Sample Dissolution

A basic aspect of sample analysis is getting the plutonium into solution. This not only applies to soil samples, but to vegetation and biological samples. Butler et al. (1971) report results of leaching techniques on two soil samples, one spiked with plutonium-238 (possibly plutonium-239), and one contaminated by an accidental release of plutonium-239. These results are summarized in Table 21.

TABLE 21. SOIL LEACHING EXPERIMENT
(From Butler, et.al. 1971)

Leach Solution	Sample Activity Based on Fusion Analysis (dpm/g)	Percent of Activity in Leached Fraction (a)
Water	1700--Spiked with ^{238}Pu, heated to 550°C	0
4N HCl	1700--Spiked with ^{238}Pu, heated to 550°C	92
12N HCl	1700--Spiked with ^{238}Pu, heated to 550°C	92
1N HF	1700--Spiked with ^{238}Pu, heated to 550°C	0
28N HF	1700--Spiked with ^{238}Pu, heated to 550°C	9
4N HCl, 1N HF	1700--Spiked with ^{238}Pu, heated to 550°C	43
4N HCl	0.57 ± 0.40--soil (b) contaminated with ^{239}Pu, heated to 550°C	39
4N HCl, 1N HF	0.57 ± 0.40--soil (b) contaminated with ^{239}Pu, heated to 550°C	87
4N HCl, 2N HF	0.57 ± 0.40--soil (b) contaminated with ^{239}Pu, heated to 550°C	> 100

a. One-gram sample boiled in 10-ml volume of leach solution, and allowed to digest for 1 hour. Results based on average of two samples. The two samples varied by less than about 10% except for the 28N HF, in which case they differed by a factor of 2.75.

b. Based on analysis of 21 1-g samples. Leach samples are based on treatment of 20-g samples with 200 ml of solution.

Table 22 indicates a comparision of leaching and fusion results from Bishop et al. (1971) of Mound Laboratory and a result from a cross check between laboratories reported by Sill and Hindman (1974). Table 23 reports data from Sill and Hindman (1974) for several different leaching conditions for plutonium fired at various temperatures.

Chu (1971) reports results for three sets of samples analyzed by the leach technique (HCL/HNO_3) and sodium carbonate fusion for both plutonium-238 and -239 (six results). The ratio of the leach:fusion results varied from 0.74 to 1.46, with four of the values being below one. The mean of the ratio was 1 ± 0.24 (1 sigma). It should be noted that Sill et al. (1974) and Butler et al. (1971) report difficulties (e.g., incomplete recovery) for sodium carbonate fusions.

Essington (1973) reports data for analysis of a soil sample spiked with plutonium (any heat treatment not indicated). The results indicate that the former LASL acid leach technique (10-g sample) only recovered about 64 percent of the plutonium-239,based on the EPA/NERC-LV method (Johns, 1975) and the modified LASL leach procedure (includes HF and $NaHSO_3$).

Majors et al. (1974, p. 107) discuss the solubility of plutonium-239 and americium-241 associated with desert vegetation. It is not certain how the plutonium is bound to the vegetation. Although some may be taken up systemically, the major fraction appears to be particulate material deposited on the vegetation (Romney, verbal presentation, NAEG, May 1975). Thus, the majority of the plutonium on vegetation probably is in the same form as plutonium in soil, although possibly associated with less siliceous material. The fraction of plutonium remaining in vegetation ash after an acid leach is given in Table 24.

The fraction of plutonium removed from soil (apparently also vegetation and probably air) samples by acid leaching is variable. Values from various investigators range from less than 50 percent to 100 percent. The validity of the 100 percent value may be questioned, since it is not based on the analysis of the leach residual. Also, it was based on a sodium carbonate fusion versus the technique of Sill et al. (1974).

In summary, it appears acid leaching may recover only roughly 60 percent of soil-related plutonium, depending on the source term. Acid dissolution, using HF may recover all of the plutonium, but prudence would indicate that samples and/or residuals should be checked by Sill's et al. (1974) pyrosulfate fusion technique.

TABLE 22. LEACHING VERSUS FUSION OF SOIL SAMPLES

Technique	Sample Activity (dpm/g)	Percent Activity Leached From Sample	Percent of Activity Found in Residue
HNO_3 leach of Pu-239 sample heated to 1000°C	35	17	81
(Sill and Hindman, 1974), Bishop et al. (1971).	35	24	78
Pu-238 in soil (a)	0.04 ± 0.008	97.5 ± 18	--
Bishop et al. (1971) (b)	0.17 ± 0.03	114 ± 36	--
Bishop et al. (1971) (b)	1.59 ± 0.31	103 ± 15	--
Bishop et al. (1971) (c)	26 ±22	53 ± 18	∿7
Sill and Hindman (1974)	35	27 (3 determinations)	No HF used

(a) Error based on 8 replicate analyses, 1-sigma error

(b) Error based on 6 replicate analyses, 1-sigma error

(c) Error based on 11 replicate analyses, 1-sigma error

The residual of 8 of the aliquots were checked for plutonium-238. The small amount of material found in the leach residual indicates that the sample activity estimate, 26 dpm/g was biased or incorrect, possibly because of discrete particulate material, and the sample size for fusion; 10 g versus 20 or 50 g for leaching. The analysis of residuals infers the leaching recovery was 93 percent versus 53 percent.

Table 23. Leachability of Plutonium from Standard Soil No.3[a] (from Sill and Hindman,1974)

Heat treatment	Plutonium standard	Acid Soluble		Residue		Total	
		dpm gram	%	dpm gram	%	dpm gram	%
2 hours at 110 °C	High[b]	29.2 ± 0.5	98.0 ± 1.6	0.89 ± 0.04	3.0 ± 0.1	30.1 ± 0.5	101.0 ± 1.6
	Low[b]	0.452 ± 0.018	89.9 ± 3.6	0.024 ± 0.004	4.8 ± 0.8	0.476 ± 0.018	94.7 ± 3.4
1 hour at 700 °C	High[b]	19.0 ± 0.3	63.8 ± 1.0	11.4 ± 0.3	38.3 ± 1.0	30.4 ± 0.4	102.1 ± 1.4
	Low[b]	0.256 ± 0.013	50.9 ± 2.5	0.246 ± 0.012	48.9 ± 2.3	0.502 ± 0.018	99.8 ± 3.5
4 hours at 1000 °C	High[b]	5.8 ± 0.1	19.5 ± 0.3	23.4 ± 0.2	78.5 ± 0.7	29.2 ± 0.3	98.0 ± 0.8
	Low[b]	0.071 ± 0.005	14.1 ± 0.9	0.422 ± 0.013	83.9 ± 2.5	0.493 ± 0.014	98.0 ± 2.7
4 hours at 1000 °C	High[c]	17.6 ± 0.2	59.1 ± 0.7	12.0 ± 0.4	40.3 ± 1.3	29.6 ± 0.5	99.4 ± 1.6
	Low						
4 hours at 1000 °C	High[d]	15.4 ± 0.1	51.7 ± 0.4	14.2 ± 0.3	47.7 ± 1.0	29.6 ± 0.3	99.4 ± 1.1
	Low[d]	0.281 ± 0.015	55.9 ± 2.9	0.224 ± 0.011	44.5 ± 2.1	0.505 ± 0.019	100.4 ± 3.6
4 hours at 1000 °C	High[e]	19.2 ± 0.2	64.4 ± 0.7	9.9 ± 0.2	33.2 ± 0.6	29.1 ± 0.3	97.6 ± 1.0
	High[e]	18.5 ± 0.2	62.1 ± 0.7	10.9 ± 0.2	36.6 ± 0.6	29.4 ± 0.3	98.7 ± 1.0

[a] Calculated values are 29.8 ± 0.1 and 0.503 ± 0.003 dpm gram of ^{239}Pu for the high and low standards, respectively. [b] Ten grams of soil was boiled for 2.5 hours with 100 ml of aqua regia. [c] Ten grams of soil was simmered in a platinum dish for 2 hours with 95 ml of concentrated nitric acid and 5 ml of 48% hydrofluoric acid. [d] Ten grams of soil was moistened with concentrated nitric acid and evaporated to dryness with 40 ml 48% hydrofluoric acid in about 1 hour. [e] Ten grams of soil heated to near boiling for 16 hours with 100 ml of either 95-to-5 or 50-to-50 of concentrated hydrofluoric acid and 8M nitric acid.

TABLE 24. PLUTONIUM LEFT IN VEGETATION ASH AFTER ACID LEACHES*
(From Majors et al., NVO-142, 1974)

Plutonium in Leach (dpm/g ash)	Plutonium in Residue (dpm/g ash)	Plutonium in Residue (Percent of Total)
118	22	16
47	19	29
151	37	20
44	0	0
63	3	5
231	66	24
354	249	41
69	3	4
54	0	0
174	20	10
284	5	2
326	0	0
355	6.2	2

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* Leached with HNO_3-HCl and H_2O_2.

ANALYTICAL VARIATION AND REPRODUCIBILITY

Gilbert and Eberhart (1974) present data on within-lab replicate sample variation. The samples are from the NAEG NTS program. Thus, some of the variation probably is due to the discrete particulate nature of the plutonium in the soil. This data reflects a range in the coefficient of variation (sigma divided by the mean) for replicates of 0.23 to 0.93.

Butler et al. (1971) report plutonium cross-check results from aliquots of five standard soil samples. Three aliquots of each sample generally differed by less than 10 percent. The individual values and their means generally were within 10 percent of the known values. The fusion method, after Sill et al. (1974), was used for analysis of the 5-g samples.

Butler et al. (1971) also report the analysis of 21 one-gram replicates of an environmental sample. The sample was taken near a nuclear facility about one year after an accidental particulate release. The contaminated area had been covered with about 12 inches of fresh dirt during the year prior to sample collection. The sample was dried, muffled at 550°C, and thoroughly mixed prior to taking the one-gram aliquots. The fusion results indicated a range of 0.25 to 1.72 dpm/g with a mean of 0.57 ± 0.40 (1 sigma) dpm/g. As indicated in Table 21, acid dissolution of 20-g samples with 4N HCl and 2N HF actually indicated slightly higher and more uniform results.

Chu (1971) and Krey and Hardy (1970) report interlaboratory results related to the HASL Rocky Flats study. The sample sets include aliquots of two samples which essentially should have been zero and several interlaboratory comparisons of different techniques. These data are given in Table 17.

Bishop et al. (1971) reports seven replicate analyses of a soil sample by the fusion technique. The sample was prepared by Sill, after the methods of Sill and Hindman (1974).

Sill and Hindman (1974) report data on an interlaboratory cross-check of their standard soil. This group of data include a comparison of duplicate analyses from seven laboratories by different techniques.

Data from AEC (1973) for the Enewetak cross-check calibration program include interlaboratory analysis of coral soil. There are five groups of data.

These various sets of data, including similar data from Table 12 are summarized in Table 25.[a] The data include the number of samples, sample size, the mean of the results, ($\bar{x}$), the standard deviation, or error, based on the averaging of the results (S), the coefficient of variation, (CV), (the standard deviation divided by the mean), and the coefficient of variation at the 95 percent confidence level (CL) based on the t-distribution (e.g., multiplied by 3.182 for 3 degrees of freedom). The actual percent analytical error also is presented ($\bar{x}-\mu/\mu$), that is, the difference of the mean from the standard value divided by the standard. Missing data are indicated by horizontal lines. The data are based on analyses of duplicate samples made up from spiked samples, and analyses of actual environmental samples. The emphasis on data selection was to use data sets illustrating analytical variability versus sampling or aliquoting variability. However, selection of data sets for this intent is admittedly subjective. The data from Bliss (1974) and Gilbert and Eberhardt (1974) probably largely reflect sample inhomogeneity versus analytical variations. The data from Bliss (1974) illustrate the reduction in result variability with the increase in sample size.

The data from Sill and Hindman (1974) come from two sources. The first two entries are from an interlaboratory calibration test using the standard soil. The other entries are from efforts to determine analytical sensitivity and sample homogeneity. The samples are based on various dilutions of the standard soil with uncontaminated soil.

Several observations can be drawn from the data in Table 25. There is a large range in the coefficient of variation (at 95 percent confidence level) in the various sets of data; it ranges from as low as 1 to 2 percent for Sill and Hindman's (1974) evaluation of the variance of analysis of standard soils, to hundreds of percent for duplicate analyses of 1-gram aliquot sizes of soils near NTS or interlaboratory analysis of soils with close to zero plutonium levels (Krey and Hardy, 1970). The nominal minimum 95 percent CV is about 10 percent where values of up to 30 to 40 percent are common.

The counting error reported by the various authors (not shown) generally is much lower than the sample result CV. Although the values were similar for Sill and Hindman's (1974) sample variance studies, the sample averaging CV generally was a factor of two or more greater than the counting CV.

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[a] The actual data sets can be obtained from the respective references or a request to Mr. David Bernhardt, Environmental Protection Agency, Office of Radiation Programs, Las Vegas Facility, P. O. Box 15027, Las Vegas, Nevada 89114.

TABLE 25. SUMMARY OF ANALYTICAL VARIABILITY OR REPRODUCIBILITY

Reference	Analytical Technique	Comments	Plutonium-239							Plutonium-238						
			Number of Samples	Sample Size (grams)	Mean, $\bar{x}$ (dpm/g)	Error, S (dpm/g)	Analytical Error, $\bar{x}-\mu_A$ (per cent)	CV S/$\bar{x}$ (per cent)	CV at 95% CL, St/$\bar{x}$ (per cent)	Number of Samples	Sample Size (grams)	Mean, $\bar{x}$ (dpm/g)	Error, S (dpm/g)	Analytical Error, $\bar{x}-\mu_A$ (per cent)	CV S/$\bar{x}$ (per cent)	CV at 95% CL, St/$\bar{x}$ (per cent)
Sill and Hindman (1974)	Fusion	Triplicate analyses by Idaho Falls lab	3	10	35.2	0.12	1.2	0.33	--	3	--	0.55	0.056	5.5	10	43
"	Mixed	Triplicate analyses by 7 labs; 1 excluded	21	0.5-10	34.1	1.19	2.0	3.5	7.3	15	--	1.01	0.44	75	43	92
"	Fusion	Analysis by Idaho Falls Lab	6	10	32.6	0.24	0.36	0.74	1.9	6	--	0.51	0.02	0.6	3.2	8.1
"	"	"	9	10	3.64	0.057	5.9	9.0	20.7	--	--	--	--	--	--	--
"	"	"	5	10	29.5	0.11	0.94	0.37	1.0	5	--	0.44	0.026	4.8	5.6	16.4
"	"	"	6	1	29.1	0.51	2.4	1.8	4.5	6	--	0.46	0.070	0.7	15	40
"	"	"	6	10	29.4	0.26	1.7	0.9	2.3	6	--	0.46	0.15	1.1	3.3	8.6
"	"	"	9	10	0.503	0.0088	0.2	1.7	4.0	--	--	--	--	--	--	--
"	"	"	6	1	0.553	0.043	10.0	7.7	19.9	--	--	--	--	--	--	--
Sill (1971)	Fusion	Ambient Soil by Idaho Falls	6	10	0.11	0.005	--	5.02	12.9	--	--	--	--	--	--	--
"	"	"	4	10	0.057	0.0059	--	10.3	32.8	--	--	--	--	--	--	--
AEC (Enewetak), (1973)	Leach	3-4 interlab analysis of coral soil	5	10-50	17.16	1.57	--	9.2	25.4	--	--	--	--	--	--	--
"	"	"	4	"	0.51	0.062	--	12.1	38.4	--	--	--	--	--	--	--
"	"	"	3	"	0.49	0.047	--	9.9	41.8	--	--	--	--	--	--	--
"	"	"	5	"	2.09	0.66	--	31.6	87.7	--	--	--	--	--	--	--
"	"	"	4	"	0.45	0.57	--	12.7	40.4	--	--	--	--	--	--	--
"	"	5 - Lab; standard solution	8	--	1278	39	--	3.1	7.3	--	--	--	--	--	--	--
Gilbert and Eberhardt (1974)	Acid dissolution	Analyzed by LASL	34	--	--	--	--	69	135	--	--	--	--	--	--	--
"	"	"	9	--	--	--	--	81	187	Probably reflect discrete particulate matter and sample inhomogeneity						
"	"	"	15	--	--	--	--	93	200							
"	"	"	15	--	--	--	--	58	124							
"	"	"	23	--	--	--	--	59	122							
"	"	"	12	--	--	--	--	26	57							
"	"	"	5	--	--	--	--	33	92							
Butler et al. (1971)	Fusion	(Analyzed by EPA(EERL)	3	5	15.77	0.15	0.55	0.97	4.2	3	--	0.34	0.055	32	16	69
"	"	Cross check samples	3	5	0.031	0.001	0	3.2	13.9	--	--	--	--	--	--	--
"	"	"	3	5	2.43	0.13	8.6	5.1	22.1	--	--	--	--	--	--	--
"	"	"	3	5	16.74	0.65	4.8	3.9	17	--	--	--	--	--	--	--
"	"	"	3	5	0.50	0.04	5.7	8.1	35	--	--	--	--	--	--	--
Chu (1971)	Leach or Na_2CO_3 Fusion	Interlab Comparison	3	--	342	50	--	15	64	3	--	6.57	0.65	--	10	43
"		of Leach & HF Dissolution	3	--	1612	15	--	1	4	3	--	33.6	3.2	--	9.4	41
"	"	and Na_2CO_3 Fusion	3	--	8.07	2.10	--	26	112	3	--	0.18	0.04	--	22.9	98
"	"	"	2	--	0.63	0.16	--	26	331	--	--	--	--	--	--	--
"	"	"	2	--	1.7	0	--	--	0	2	--	0.23	0.26	--	116	1476
"	"	"	4	--	0.42	0.06	--	13.8	44	4	--	0.11	0.11	--	100	318
"	"	"	4	--	4.5	0.29	--	6.4	20	4	--	0.17	0.12	--	70	223
Bishop et al. (1971)	Fusion	Mound Lab-Soil Std.	--	--	--	--	--	--	--	6	1	36.8	2.43	1.1	6.6	17
Krey & Hardy (1970)	Leach	Pre-1945 Sample; 3 Labs	5	--	0.004	0.009	--	215	598	5	--	0.0012	0.0024	--	200	555
"	"	Excludes anom. from above	4	--	0.00015	0.0001	--	67	212	4	--	0.00013	0.00005	--	40	127
"	"	Sample from below 90 cm.	5	--	0.014	0.018	--	136	377	5	--	0.00028	0.00035	--	125	346
"	"	Excludes anomolous result from above	4	--	0.0054	0.0013	--	23	74	--	--	--	--	--	--	--
Bliss (1973) see Table 10)	Acid dissolution	Aliquots of EPA	14	1	1.095	1.419	--	130	280	Data treatment in Table 10 by log normal. Range of data probably related to discrete particulate nature of Plutonium contamination						
"	"	a single Rielo	10	10	2.326	1.53	--	66	149							
"	"	sample from LLL	4	25	3.325	1.109	--	33	106							
"	"	near NTS LLL	4	100	5.025	1.198	--	24	76							
Robinson et.al. (1975)	"	Replicates of Background	--	--	--	--	--	--	--	50	10	0.077	0.123	--	160	320

Figures 5 and 6 are histograms of ratios of results from the AEC (1973) Enewetak program. The ratios represent the results for samples split between the Air Force McClellan Central Laboratory (MCL) and either the Eberline Instrument Company (EIC) or the Laboratory for Electronics, Environmental Analysis Laboratories Division (LFE).

The histograms of the data indicate that the LFE/MCL ratios (Figure 5) are centered around one. AEC (1973) concluded, based on a log normal treatment of the data (geometric mean of 1.02) that the average was not statistically different from zero (95 percent confidence).

The data in Figure 6 indicate the EIC/MCL data are centered about 0.8. The geometric mean, excluding the lowest value of less than 0.1, is 0.85, and indicates statistically significant bias.

The interlaboratory calibration indicated that the EIC value was 94 percent of the average of the other laboratories and 95 percent of the average of MCL. The LLL:MCL ratio was 0.96. The calibrations of these laboratories may not have all been independent. The intent of this discussion is to indicate the variations, not which laboratories were correct.

Robinson et al. (1975) report data from 20 samples split between Mound Laboratory and EPA/EERF. These data indicated a Mound:EPA average ratio of 0.93 ± 0.12 (1 sigma).

Table 26 contains data extracted from Table 25 which are related primarily to analytical variability. The values have been catagorized by the relative level of activity in the sample. Table 27 includes data that generally are related to both sampling and analytical variability.

The lowest sample value in Table 26 (Krey and Hardy, 1970, 0.00015 pCi/g) indicates the greatest variation. This is indicative of analysis near the MDA. The one-sigma counting error for these results was 100 percent of the mean. Assuming a 100-gram sample and roughly 75 percent tracer yield, the counted sample would have contained about 10 fCi (0.00015 pCi/g x 10^3 fCi/pCi x 100 g x 0.75). This sample should have been zero, since it was collected prior to 1945. The value of slightly above zero may relate to minor sample handling or analytical contamination (data given in Table 17).

The data from Robinson et al. (1975) reflect the variability of low level results and the importance of instrument and reagent blank background. The results are based on gross counts with no background subtraction.

Other than the previously discussed "zero" sample in Table 26, the coefficient of variation values show a limited

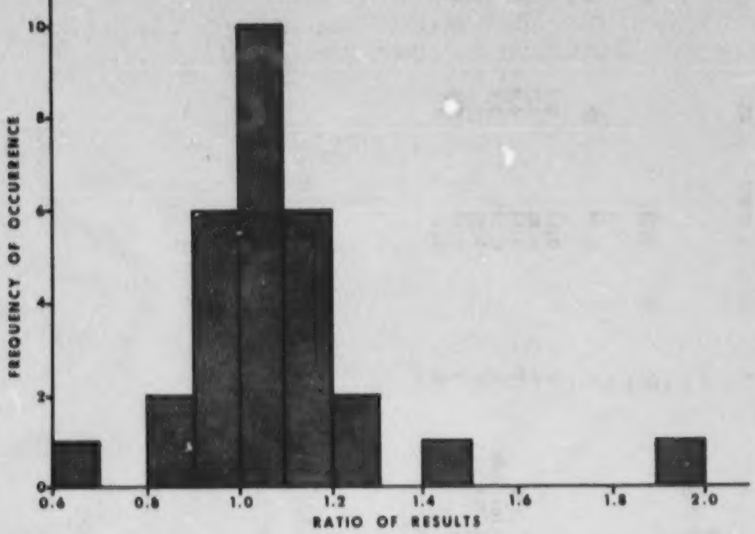

Figure 5. Histogram of ratio of duplicate soil sample results (LFE/MCL) from Enewetak, (Data from AEC,1973))

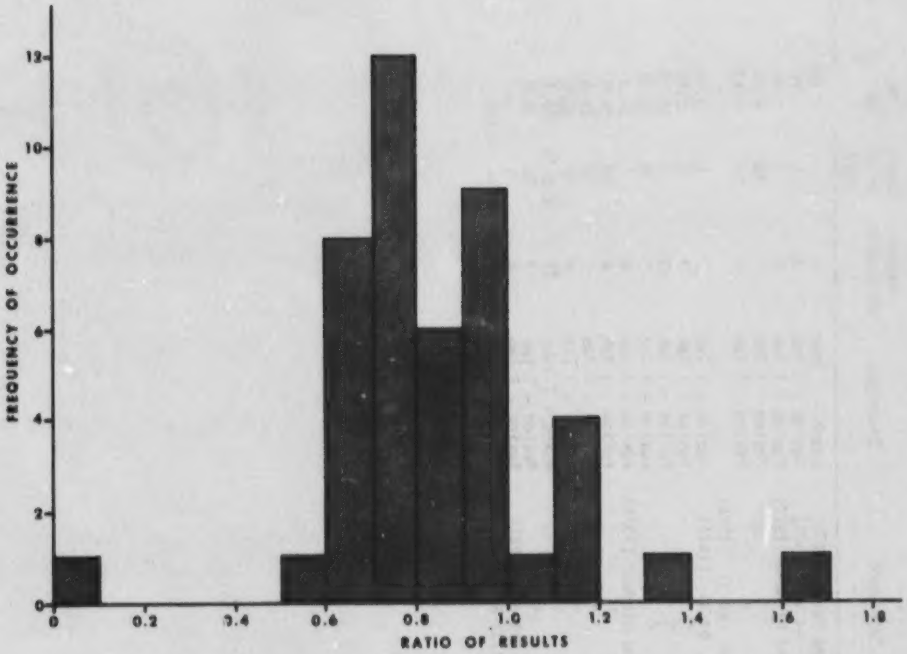

Figure 6. Histogram of ratio of duplicate soil sample results (EIC/MCL) from Enewetak, (Data from AEC,1973)

TABLE 26. VARIABLIITY OF ANALYTICAL RESULTS

		Plutonium-239						Plutonium-238				
Reference	Technique	Number of Samples	Sample Size (g)	Mean (pCi/g)	Difference From True Value (per cent)	CV at 95% CL (per cent)		Number of Samples	Sample Size (g)	(pCi/g)	Difference From True Value (per cent)	CV at 95% CL
Krey and Hardy (1970)	Leach - 3 labs	4	--	0.00015	67	212		4	--	0.00013	40	127 %
Bulter et al. (1971)	Fusion - 1 lab	3	5	0.031	0	14		--				
Sill and Hindman (1974)	Fusion - 1 lab	6	1	0.55	10	7.7	11±9%	--				
"	Fusion - 1 lab	9	10	0.50	0.2	4.0		--				
"	Fusion - 1 lab	9	10	0.64	5.9	21		--				
Robinson et al. (1974)								50	10	0.077	--	320 %
Bulter et al. (1971)	Fusion - 1 lab	3	5	0.50	5.7	35		--				
"	Fusion - 1 lab	3	5	2.43	8.6	22		--				
"	Fusion - 1 lab	3	5	15.77	0.6	4.2		3		0.34	32	69 %
"	Fusion - 1 lab	3	5	16.34	4.8	17		--				
Sill and Hindman (1974)	Fusion - 1 lab	6	1	29.1	2.5	4.5	2±2% (exclude 7%)	6		0.46	0.7	40 %
"	Fusion - 1 lab	6	10	29.4	1.7	2.3		6		0.46	1.1	8.6%
"	Fusion - 1 lab	5	10	29.5	0.9	1.0		5		0.44	4.8	16 %
"	Fusion - 1 lab	6	10	32.6	0.36	1.9		6		0.51	0.6	8.1%
"	Mixed - 7 labs	21	0.5-10	34.1	2.0	7.3		15		1.01	75	92 %
	Fusion - 1 lab	3	10	35.2	1.2	0.33		3		0.55	5.5	43 %
Biship et al. (1971)	Fusion - 1 lab	6	1	--				6		36.8	1.1	17 %
AEC (1973) Enewetak	Leach - 5 labs	3	--	1276.	3.1	7.3		--				

TABLE 27. VARIABILITY OF ENVIRONMENTAL SOIL SAMPLE RESULTS

Reference	Technique	Type Sample	Plutonium-239 Number of Samples	Sample Size (g)	Mean (pCi/g)	CV at 95% CL	Plutonium-238 Number of Samples	Sample Size (g)	Mean (pCi/g)	CV at 95% CL
Krey and Hardy(1970)	Leach-3 labs	Pre-1945	4	--	0.00015	212%	--	--	0.00013	127%
Krey and Hardy(1970)	Leach-3 labs	"(above + 1)	5	--	0.004	598%	--	--	0.0012	555%
Krey and Hardy(1970)	Leach-3 labs	Sample below 90 cm	5	--	0.014	377%	--	--	0.00028	346%
Krey and Hardy(1970)	Leach-3 labs	"(above - 1)	4	--	0.0054	74%	--	--	0.00028	346%
Butler et al.(1971)	Fusion	Amb. Soil	21	--	0.57	144%	--	--		
Robinson et al.(1975)		Background					50	10	0.077	320%
Chu(1971)	Mixed	Amb. Soil	4	--	0.42	44%	--	--	0.11	318%
Chu(1971)	Mixed	Amb. Soil	2	--	0.63	331%	--	--	--	
Sill(1971)	Fusion-1 lab	Ambient Soil	4	10	0.057	33%	--	--		
Sill(1971)	Fusion-1 lab	Ambient Soil	6	10	0.11	13%	--	--		
AEC(1973)Enewetek	Leach-3 or 4 lab	Coral Soil	4	10-50	0.45	40%	--	--		
AEC(1973)Enewetek	Leach-3 or 4 lab	Coral Soil	4	10-50	0.49	42%	--	--		
AEC(1973)Enewetek	Leach-3 or 4 lab	Coral Soil	4	10-50	0.51	38%	--	--		
Bliss(1973)	Accid Dissol	Near NTS	14	1	1.095	280%	Aliquots of same sample,			
Bliss(1973)	Accid Dissol	Near NTS	10	10	2.326	149%	probably reflect discrete particulate			
Bliss(1973)	Accid Dissol	Near NTS	4	25	3.325	106%	non-homogeneous nature of NTS			
Bliss(1973)	Accid Dissol	Near NTS	4	100	5.025	76%	related Pu contamination			
Chu(1971)	Leach or Fusion	Amb. Soil	2	--	1.7	0%	--	--	0.23	1476%
Chu(1971)	Leach or Fusion	Amb. Soil	4	--	4.5	20%	--	--	0.17	223%
AEC(1973)Enewetah	Leach 3 or 4 lab	Coral Soil	5	10-50	2.09	89%	--	--		
Chu(1971)	Leach or Fusion	Amb. Soil	3	--	8.07	112%	--	--	0.18	98%
AEC(1973)Enewetah	Leach 3 or 4 lab	Coral Soil	5	10-50	17.16	25%	--	--		
Chu(1971)Rocky Flats	Leach or Fusion	Amb. Soil	3	--	342	64%	--	--	6.57	43%
Chu(1971)Rocky Flats	Leach or Fusion	Amb. Soil	3	--	1612	4%	--	--	33.6	41%
Eberhart & Gilbert(1974)	Accid Dissol lab	NTS Soil	34	--	--	135%				
Eberhart & Gilbert(1974)	Accid Dissol lab	NTS Soil	9	--	--	187%	Probably reflect discrete			
Eberhart & Gilbert(1974)	Accid Dissol lab	NTS Soil	15	--	--	200%	particulate nature of			
Eberhart & Gilbert(1974)	Accid Dissol lab	NTS Soil	15	--	--	124%	NTS related Pu contamination			
Eberhart & Gilbert(1974)	Accid Dissol lab	NTS Soil	23	--	--	122%				
Eberhart & Gilbert(1974)	Accid Dissol lab	NTS Soil	12	--	--	57%				
Eberhart & Gilbert(1974)	Accid Dissol lab	NTS Soil	5	--	--	92%				

relationship to relative levels of sample activity. Observing only Sill and Hindman's (1974) data, the values below 1 pCi/g appear to have about five times the variation of the values above 29 pCi/g average of 11 ± 9% versus 2 ± 2% at 1 sigma). The respective geometric means and geometric standard deviations are $\bar{x} = 9\%$, $s = 2.3$ and $\bar{x} = 1.5$, $s = 2.7$ respectively. However, this is a limited amount of data (only two concentration classes) from which to make a conclusion. There are not sufficient results from interlaboratory studies to conclude that they have more variance than intralaboratory studies, although there is an indication of this.

The data indicate more uncertainty in the plutonium-238 results than in the plutonium-239 results. Excluding the "zero" result, the means of the percentage uncertainties for concentrations around 1 pCi/g or less are 37 ± 31 percent for plutonium-238 versus 11 ± 9 percent for plutonium-239.

The first four entries in Table 27 are for the above-mentioned pre-1945 sample, and a sample collected in 1970, at a depth of 90 cm. Both samples were analyzed by three laboratories. In both instances the same laboratory presented results about an order of magnitude above the other laboratory's results. Upon request, the sample was re-analyzed with lower, but still elevated, results. The disparity is the reason for the assessment for both four and five samples.

It is possible that the soil sample from 90 cm was contaminated by natural movement of plutonium. However, Krey and Hardy (1970) note that it is probably more likely that the sample was contaminated during collection and handling. This implies the difficulties of handling and collecting samples of grossly different levels of contamination without minor cross contamination occurring. The associated problems are the basis for the recommendation that samples of various stratified activity levels be collected and analyzed separately. Minor cross contamination from one sample can grossly affect the results of a sample of much lower activity.

SUMMARY AND CONCLUSIONS

This report has considered both field instrumentation and sampling and analyses techniques for assessing environmental plutonium concentrations. The report has centered primarily on soil and air sampling techniques and plutonium-238 and -239 analytical techniques. However, much of the information applies to the transuranic elements in general and to other types of samples.

Field instrument techniques are not sensitive enough to assess the ambient environmental levels (roughly 1 nCi/m^2 or less than 1 pCi/g in soil). The FIDLER's sensitivity is indicated as about 130 nCi/m^2, but the variabilities associated with field work indicate uncertainties at even 200 to 500 nCi/m^2 (roughly 50 pCi/g), and the need for confirmatory radiochemistry analyses. Several refinements can be made in using the FIDLER, but basicially it is a survey instrument, not an instrument for quantitating concentrations.

There are several photon counting techniques that allow direct estimation of americium-241, and to a limited extent, plutonium-239 (X-rays), with associated estimates of plutonium-239. The general sensitivities range roughly from 1 pCi/g for americium-241 to 20 pCi/g for plutonium-239.

There are six basic sources of error or variation in relation to plutonium and other transuranic analysis of environmental samples. These are sampling technique (soil), sample size, sample dissolution, inadequate chemical equilibrium between sample plutonium and the tracer, interfering elements, and quantitation of results.

SAMPLING TECHNIQUE (SOIL)

Items to be considered in sampling programs include:

1. Sample representative of stated conditions; i.e., stated depth and area for soil samples. The area sampled should be sufficient to account for minor inhomogeneity.
2. Sample of pertinent depth; i.e., adequate depth to measure total inventory (if that is the objective) and appropriately limited depth to prevent unnecessary

dilution of contaminated layer for deposition or resuspension studies.

3. Generally, it is recommended that samples should represent an area of about 1000 cm^2 (1 ft^2). The variance associated with this or smaller areas has not been quantitated, and would be source-dependent. Sampling errors for a sample of 1-cm depth or less are estimated to be up to 50 to 100 percent. The sampling error for a 5-cm depth (100 cm^2) are estimated to be limited to about 20 percent. The estimate of 20 percent is based on soil mechanics theory (Terzaghi and Peck, 1968) and a ORP-LVF field experiment.

SAMPLE SIZE

The potential for plutonium contamination to exist as discrete particles results in a potential variation in sample results of up to several orders of magnitude (roughly 95 percent C.L.) depending on the sample size analyzed and the particle size of the plutonium contamination. The ratio of the upper and lower limits, at the 95 percent confidence level, for 1-gram aliquots of samples is roughly a factor of 10 or more, based on a log-normal distribution (see Table 11). It appears this ratio may be reducible to about 2 (Sill, 1971 data, Table 9) by using 10-gram samples. The ratio is reduced further, at least for less homogeneous samples, by using 25-gram and 100-gram samples.

Michels (1971) evaluated two groups of data for the Rocky Flats, Colorado area. Using a log-normal distribution, he divided the data of Krey and Hardy (1970) into two distributions. One, for global fallout, had a geometric mean of about 2 nCi/m^2; the other, generally relatable to Rocky Flats contamination, had a geometric mean of about 15 nCi/m^2. He notes that the data of Poet and Martell (1972) range somewhat lower (units of nCi/m^2) than those of Krey and Hardy. This probably is due to the more shallow sample depth, 1 cm, versus 20 cm for Krey and Hardy (1970). Furthermore, the data cannot be split reasonably into the two distributions, possibly because of the increased variance associated with shallow sample depth and small sample aliquot size (10-g versus 100-g for Krey and Hardy).

Particle Size Distribution of Plutonium In Soil

Soil samples from Rocky Flats, Colorado, were partitioned into size categories of less than 2 mm, 100 micrometer, and 10 micrometer diameters. The mass fractions based on the soil less than 2 mm in diameter were 43 percent and 20 percent, for the less than 100-micrometer and 10-micrometer (density 1 g/cm^3) diameter partitions, respectively. The plutonium-239 concentrations (pCi/g) for the 100-and 10-micrometer fractions were 1.8 and 2.5, respectively, times the concentration in the less than 2 mm fraction. These results are in general agreement with those of other investigators.

SAMPLE DISSOLUTION

Sill et al. (1974) and Sill and Hindman (1974 indicate that non-fusion techniques may leave up to 40 percent of refractory plutonium in the undissolved residual for siliceous soils. Butler et al. (1974) further note that sodium carbonate fusions, etc. are not as successful as Sill's et al. (1974) potassium fluoride and pyrosulfate fusions. Furthermore, it is not sufficient to get plutonium into solution only, but it must also be in the monomeric ionic state. Failure to obtain proper chemical equilibrium (ionic state) between the sample plutonium and the tracer gives invalid results, which may be either high or low.

Although acid leaching is adequate in some instances (Krey and Hardy, 1970, and Chu, 1971), it may recover only up to about 60 percent of the plutonium in siliceous soils. Acid dissolution with HF appears to recover more of the plutonium, but still has a potential for incomplete recovery. It would appear to be prudent to check insoluble residuals and complete samples with Sill's et al. (1974) fusion technique. Hahn et al. (in press) and Bretthauer et al. (1975) have proposed this technique as the tentative EPA standard method.

INTERFERING ELEMENTS

Various elements (e.g., fluorine, calcium, iron, uranium, etc.) can interfere with the various separation techniques and stages in the preparation of actinide samples for counting. It is common, and would appear to be necessary, to use tracers (e.g., plutonium-236 or -242) for radiochemistry determinations (other tracers for other elements). Thus, although all of the sample plutonium may not be recovered, the fraction lost, other than from incomplete dissolution, generally is accounted for. Thus, any inaccuracies result in low (lack of dissolution of sample plutonium) results. However, there are two additional considerations. If the plutonium tracer is placed in an empty beaker, it may bond to the beaker and be partially lost. The sample plutonium would be recovered nominally. Furthermore, some techniques (spills and bubbling or spattering) may result in some loss of tracer prior to dissolution of the sample plutonium.

The second consideration is the effect on analytical sensitivity of low tracer yields. This is difficult to quantitate, but a yield of 10 percent has more potential uncertainty than a yield of 100 percent. Plus there is the increased counting error associated with the lower count rates of low recoveries.

Sill et al. (1974) note that the electrodeposition step is the point of greatest potential plutonium loss (reduced yield) in the procedure for alpha pulse height analysis. This is due partially to the potential for plutonium co-precipitation with other elements or the general formation of insoluble or non-electroplatable plutonium. There is the additional problem of uranium or other elements being included in the electroplating and forming a non-weightless plate, with associated alpha self-absorption. Although liquid scintillation counting and co-precipitation of plutonium with trace amounts of lanthanum present alternatives to electrodeposition, electrodeposition appears to be the technique preferred by most laboratories.

QUANTITATION OF RESULTS

The quantitation of results is done by alpha spectroscopy of an essentially weightless sample or liquid scintillation, or mass spectrometry. All of these techniques require radiochemical treatment prior to final analysis. They each have their unique interference problems. Although mass spectrometry is inherently and, at the present state of the art, more sensitive than alpha spectrometry, there is interference from hydrocarbons, and because of the high specific activity of plutonium-238 and interference from uranium-238, the sensitivity for plutonium-238 is poor. Mass spectrometry has the advantage in that it provides isotopic ratios for plutonium-239,-240, and -241 and thus can often be used to relate contamination to specific sources, even when the plutonium-239 contributions are similar.

There are several means of defining the sensitivity of analyses, or minimum detectable activity. The techniques that give the lowest MDA's that are reasonably valid are based on the two or three-sigma counting error. The NERC-LV technique (Johns, 1965), defines the MDA value as the mean value equal to the two-sigma error. Others, (Eberline) sometimes use three times the background counting error, which generally gives results similar to Johns (1975). In most instances when mean sample results are below or equal to the MDA, they are expressed as less than the MDA.

It should be recognized that most less than values are only a 50 percent probability statement. That is, 50 percent of the time the statement is wrong. A reasonable minimum MDA is about 20 fCi per sample; i.e., the counting error is 100 percent at the 2-sigma or 95 percent confidence level. In essence, these values are per sample planchet, after electroplating. If the chemical (tracer) yield is only 50 percent, the values per original sample are doubled.

Variation of Results

An indication of the variance associated with the analysis of samples and with both sampling and analysis is presented by the data summarized in Table 28. These data are summarized from the presentations in Tables 26 and 27. The variance of results is presented in terms of both normal and log-normal distributions. This is not to imply that the data fit these distributions, rather, they are used as tools to summarize the data.

The data in Table 28 illustrate that the variation of environmental soil sample results is much greater than just the analytical variation. The variance associated with sampling and analysis for samples equal to or less than 1 pCi/g is more than twice that associated with just the analysis of samples. There are mixed groups of data and the categorizations may be subjective, but the majority of the various data sets clearly indicate that sampling and analytical error, or just analytical error, exceed the counting error by several factors. Consideration of the data compiled in Tables 25, 26, and 27 indicate that variances of less than 20 percent are the exception, rather than the rule, even for results significantly above ambient concentrations.

The following points become evident:

1. Analytical results for 10-gram samples at ambient plutonium-239 levels (less than 1 pCi/g) can be expected to have (95 percent confidence) coefficient of variation of about 10 percent plus or minus a factor of two. Plutonium-238 results can be expected to have a coefficient of variation of about 30 percent, plus or minus a factor of two or three.

2. Reasonably homogeneous soil samples (10-gram) can be expected to have a 95 percent confidence level coefficient of variation (CF/95 percent) of about 50 percent plus or minus a factor of up to four (one-sigma).

3. Soil samples characteristic of NTS, presumably with discrete particulate material, appear to be characterized with a coefficient of variance of over 100 percent plus or minus a factor of about two. The size of the sample aliquot affects the variation.

4. Data reported by Bliss (1974) exemplify the decrease in variability of heterogeneous samples with an increase in the sample size that is analyzed. The CF/95 percent decreased from 280 percent to 76 percent for 1-gram and 100-gram samples, respectively. This is between a cube and fourth-root relationship.

5. Plutonium-238 soil sample results at ambient levels indicate extreme variability, although only limited data were available.

It should be recognized that the above conclusions are based on normal and log-normal treatments of the data. No tests have been made concerning the applicability of these treatments. However, the statements are not intended to be statistically-proven hypothesis, rather they are indications of trends and categorizations of the data.

TABLE 28. SUMMARY OF VARIATIONS ASSOCIATED WITH ANALYTICAL RESULTS AND SAMPLING AND ANALYSIS RESULTS

Data Averaged	Average of 95 Percent CL Coefficient of Variation (Percent)			
	Normal Distribution		Log Normal Distribution	
	Average	Standard Error	Geometric Mean	Geometric Standard Deviation
Analytical Results				
1. Plutonium-239, ≤ 1pCi/g	49	81	20	4.0
2. Plutonium-239, ≤ 1pCi/g Kreg & Hardy Excluded	16	12	13	2.3
3. Plutonium-239, ≃ 30 pCi/g	3	3	1.9	3.0
4. Plutonium-238	74	95	39	3.4
Sampling and Analysis Results				
1. Plutonium-239, ≤ 1pCi/g	110	110	72	2.8
2. Plutonium-239, Other than NTS	76	86	39	4.1
3. Plutonium-239, NTS	140	64	130	1.6
4. Plutonium-239, Bliss, NTS	150	90	135	1.8
4. Plutonium-239 Gilbert & Eberhardt (1974) NTS	130	50	120	1.5
5. Plutonium-239, all	130	190	62	3.6
6. Plutonium-238	330	440	185	3.1

There does not appear to be adequate in-depth data for a rigorous evaluation of analytical and sample result variability. Data of this type probably can be obtained through specially designed intralaboratory and interlaboratory studies.

The results from Tables 26 and 27 contribute confirmation to the previous conclusions.

The following items summarize some additional pitfalls and concerns that must be considered in evaluating environmental data:

1. Low chemical or tracer yields are indicative of difficulties or interferences in the analysis. Prudence would seem to indicate reduced confidence in low yields (roughly defined as below 50 percent, or especially 20 to 30 percent). Low yields can occur because of loss of tracer prior to dissolution of sample plutonium, etc.

2. Sample aliquoting procedures can result in nonrepresentative results. Lung, liver, kidney, etc. samples are not homogeneous. If sections are taken for analysis (especially pertinent to large bovine organs), care must be taken to take representative fractions.

3. Gonads, lymph, etc. produce very small samples, resulting in limited sensitivity.

4. Soft tissues produce variable ash weights depending on the time and temperature of ashing. Results reported per gram of ash are thus variable. Wet tissue weights also are effected by dehydration, depending on packing techniques.

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APPENDIX A

EMSL
WORKSHOP RECOMMENDATIONS
ON
SAMPLING AND ANALYSIS

Summarized by
Dr. Bernd Kahn
and
E. B. Fowler

WORKSHOP RECOMMENDATIONS ON ANALYTICAL PROCEDURE

Dr. Bernd Kahn Analysis Report April 3, 1974

It is my impression that the consensus was as follows:

1. EPA should consider two analytical procedures:

   a. The HSL (Idaho Falls AEC Health Services Laboratory) fusion method.

   b. A total dissolution with HNO_3-HF. (Several versions available, some more promising than others. For example, a Los Alamos Scientific Laboratory method which is also the AEC Regulatory Guide Method. More detailed method descriptions, error evaluations and definitions of limits are available and should be obtained from the different laboratories.)

2. In addition, references should be made in the EPA Reference Method to procedures that have special advantages, for example, for processing large samples or numerous samples. The applicability of using these latter methods should be confirmed by comparison with the above cited reference methods.

3. The proposed reference methods should be tested independently by EPA before recommending them.

4. The 10 gram sample size appears to be appropriate, but required minimum detectable levels should be arrived at by the EPA to determine if the 10 gram samples are indeed sufficiently sensitive.

5. The major contribution to the variability of results is believed to be the occurrence of "hot" particles. It is desirable that studies be undertaken to check the influence of sample size in this variability. Guidance should be presented in the Reference

Method to assure that samples will be sufficiently large to minimize variability for the particle size expected at the location from which the methods will be used.

6. Methods should include thorough discussions of the principles and purposes of each of the procedural steps; guidance for minimizing errors, identifying the sources of the errors and calculating the magnitude of the errors; and specify a quality assurance program, including a program for minimizing cross contamination.

7. The importance in using sufficient plutonium tracer for achieving high precision should be indicated. Both Pu-236 and Pu-242 are satisfactory if they are sufficiently pure.

5/74
EBF

COMPILATION FROM THE EPA APRIL WORKSHOP

SOIL SAMPLING

Discussions relating to soil sampling consisted of four parts: (1) presentation of papers, (2) panel discussion, (3) group discussion to fix objectives, and (4) a synopsis of group conclusions.

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Sampling for two general types of mission for radioisotope measurement were identified:

1. Sampling for low levels of radioisotopes such as that associated with worldwide fallout, specifically for preoperational environmental surveillance or the establishment of a base line prior to the installation of a facility; and

2. Sampling to determine levels of radioisotope dispersal due to release associated with accidental incidents, testing, or routine plant emission.

Although the above two are different in some respects, a basic sampling procedure will apply to both situations. In the above two situations there can be permutations such as an abbreviated survey to locate areas for more intensive sampling in case (1), or in case (2) an abbreviated survey to determine whether a suspected release has occurred and if it has, its possible extent.

Further, in case (1), pre and post operational surveys will be required to determine and document the effect or lack of effect of operations on the environment. Case (2) may require an inventory either immediately after or at some period of months or even years after an incident.

It can be argued that continuous air monitoring is sufficient for industrial plant environs; this may be especially true since the predicted plant of the future is a plant of "zero emission". However, since the soil is an integrator and is relatively stable with respect to air it is a desirable matrix for programs involving extended sampling.

With the above factors in mind, the following recommendations are made relative to the establishment of an on-going soil sampling program. It is recognized that some of the permutations referred to will negate certain recommendations, however, any sampling protocol, even the simplest, should fall within the boundaries set forth herein.

The boundaries which define problems associated with a sampling program are outlined in Fig. 1; the objectives to be met are listed in Fig. 2.

The objective of any sampling method is to obtain a representative sample. The following outline is set forth as a guide to accomplish that objective. In the connotation used here, sample preparation is included as a part of the sampling scheme.

Let us assume an extended sampling program which could entail four phases;

1. pre-operation or base line,
2. operational or environmental surveillance,
3. operational incident or release,
4. post release monitoring and inventory.

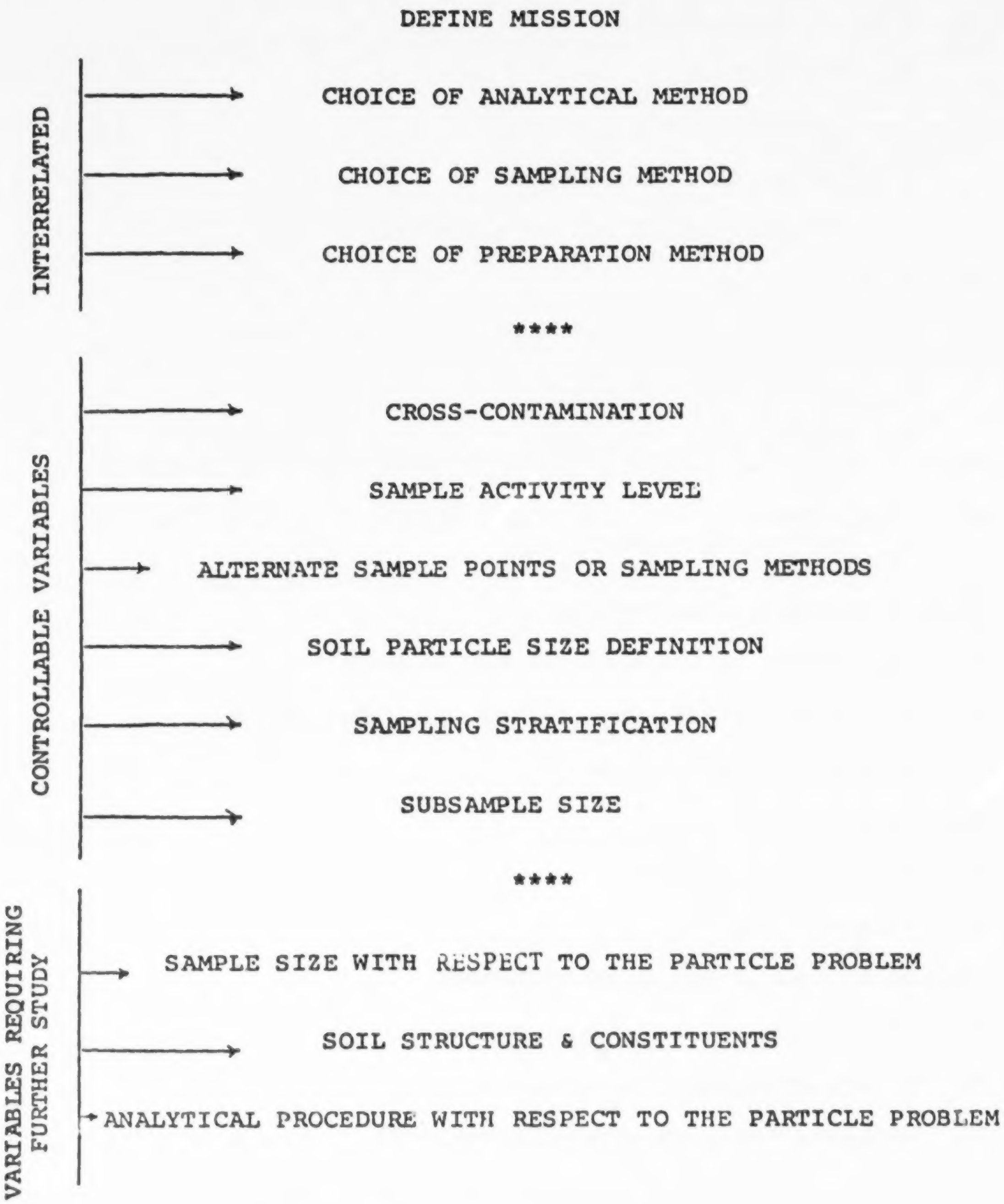
Figure 2

OBJECTIVES

- I. Define Mission
  - A. What is the purpose
    - 1. Pre-op survey
    - 2. Confirm trends
    - 3. Determine inventory
      (routine operation or accident)
  - B. What nuclides
  - C. What degree of precision is to be considered
    - 1. Number samples/location
    - 2. Number anal./sample
    - 3. Cost
    - 4. Analytical sensitivity
- II. Choice of Analytical Method
  - A. Dependence on sample type and size
  - B. Degree of confidence greater than that defined in mission definition
- III. Sampling Methods
  - A. HASL Method
  - B. NAEG Method
- IV. Choice of Sample Preparation Method
  - A. Dependence on analytical method
  - B. Define excludable material
  - D. Consider need for special treatment

BOUNDARY CONDITIONS

Figure 1


DEFINE MISSION
INTERRELATED
CHOICE OF ANALYTICAL METHOD
CHOICE OF SAMPLING METHOD
CHOICE OF PREPARATION METHOD
****
CONTROLLABLE VARIABLES
CROSS-CONTAMINATION
SAMPLE ACTIVITY LEVEL
ALTERNATE SAMPLE POINTS OR SAMPLING METHODS
SOIL PARTICLE SIZE DEFINITION
SAMPLING STRATIFICATION
SUBSAMPLE SIZE
****
VARIABLES REQUIRING FURTHER STUDY
SAMPLE SIZE WITH RESPECT TO THE PARTICLE PROBLEM
SOIL STRUCTURE & CONSTITUENTS
ANALYTICAL PROCEDURE WITH RESPECT TO THE PARTICLE PROBLEM

A basic requisite to such an extended sampling program relates to the final product, i.e. data reduced to a form that is communicable and meaningful. Statistical advise at all stages of such an extended sampling program is a must. The rationale for statistical advice is two-fold (1) communication of results on a common base,and (2) legalistics of today demand numbers which cannot be refuted.

The sampling program may be nonbiased or biased. A statistically designed program requiring random sampling would be considered nonbiased; an extended sampling program should fall into this category. An abbreviated survey might be biased and could serve an immediate "need to know". It might be acceptable under those conditions or as a starting point or base for a nonbiased extended survey.

Figures 1 and 2 outline the parameters which must be defined in an approach to an acceptable sampling procedure; they will be discussed in detail.

I. Define the Mission

A. What is the reason for the sampling program?

1. In the assumed case of an extended sampling program, the reason for sampling will change with time, however, a first requirement will be the establishment of a base line, the pre-operational survey. The survey should be both extensive and intensive in that future trends and conclusions may be based on the initial findings. A well-planned, random sampling scheme will be valuable in the determination

of an initial "inventory". Aside from test, accident, or discharge site, the concentrations of radionuclides found is expected to be low.*

2. A second purpose is that of defining or confirming trends. Any trend will be related to the base line. It is probable that a larger number of samples will be required during early operational experience and that samples will be taken at greater frequency. Complete documentation as in (1) above will prevent a resampling of previous points.

3. A third purpose is that of determining an inventory particularly associated with and after an accidental emission. In case (1) and (2) above, a surface sample only might be required, however for inventory purposes, profile samples will be necessary to assure that the highest practical percentage of a radionuclide has been accounted for.

It is recognized that resuspended soil could have been or may become a part of the soil sample, however, the separate sampling of that fraction is a special case and is not considered in this discussion.

B. What radionuclides are being sought and what is the physical nature of the dispersed material? Plutonium is considered the nuclide of prime importance, however, the sampling scheme is applicable to many radionuclides, including other transuranics as well as uranium. Radionuclides of interest in the future might include the noble gases and tritium, though proposals stated here do not consider such special cases.

---

*Drainage areas, low spots, areas of heavy vegetation, and denuded areas should be noted. The basic survey should be as completely documented as possible.

The physical nature of the dispersed material will dictate the sampling design in that particulate material widely dispersed will produce "hot" and "cold" samples whereas an emission of material in a soluble form will result in samples more nearly uniformly distributed in extent of activity. Further, the analytical method of choice may be dictated by the physical nature of the material, e.g. a refractory oxide as opposed to material deposited from solution.

The degree of solubility of the nuclide, hence transport through the soil profile will dictate the need for profile samples and their depths hence the sampling scheme should consider the chemistry of the nuclide being sought as well as the purpose for which samples are taken.

C. What is the acceptable degree of precision?

The acceptable degree of precision is probably a management decision, however, it is related to the defined purpose for sampling. Certainly the need to determine post-incident inventory would require a high degree of precision if a material balance is to be attempted.

For the following discussion, several terms should be defined; it should be pointed out that some definitions may be redundant; they are given here for explanatory reasons:

1. Sample

   That discrete mass removed from a single sampling point.

2. Sampling point

   The defined volume from which a discrete sample is removed.

3. Sampling location

   A volume delineated by a section of a grid, by an isopleth or by other means which may define a supposed commonality (of activity) to the enclosed population.

4. Sampling area.

   The area of experimental interest whose boundaries are defined by the mission. It may be a small area, say 10 x 10 feet associated with a spill or as in the case of worldwide fallout, it is the surface of the earth.

   In considering the degree of precision, the following should be factored into a decision.

   a. The number of samples taken at a location. Certain factors will dictate this number;

they may be knowledge of the problem, previous sampling results, field instrument survey, or others.

b. The number of analyses per sample. Heterogeneous distribution of particulate matter which varies in size will dictate replication to establish "within sample" variation. Once established, replication on a given percentage of samples should be practiced to confirm the presence or absence of change.

The more nearly homogeneous the sample, the fewer the replications which will be required.

c. Cost. It is obvious that the greater the number of samples taken and the greater the number of analyses performed, the more precise is the resultant number. Cost will affect the number of samples taken and the number of samples analyzed; a compromise is necessary. The extent of the compromise will be dictated by the mission.

Gilbert (1) has given the following guidance relating the above factors.

$$M_{opt} \doteq \frac{s_a}{s_\mu}\sqrt{\frac{C_1}{C_2}}$$

where: C_1 = cost of taking and preparing one sample from field

C_2 = cost of alalyzing one aliquot

s_μ = standard deviation relating to variation in the field position, and

s_a = standard deviation relating to variation within a given soil sample.

The relationship does consider cost and standard deviation and forms one basis for consideration of the degree of precision.

d. Analytical sensitivity

The adoption of a common analytical method would represent the ideal. In the absence of an ideal situation, the realistic approach is to choose only those methods of analysis which have been shown to yield comparable results and sensitivities. The mission will dictate the sensitivity required in that short cuts in a method may yield results less precise but acceptable to the mission. Such a case might arise when a few samples are taken downwind after a suspected emission as a first check to determine "whether and to what extent". Certainly high analytical sensitivity is desired in the determination of a "most practicable inventory".

II. Choice of the Analytical Method

A. The possible analytical methods of choice will be determined by the group reviewing that problem. However, the choice will be dependent on certain characteristics of the sample. It is also true that characteristics of the sample may dictate minor modifications associated with analyses. As an example, a low organic soil may be digested without pretreatment whereas a high organic soil becomes more amenable to analyses if an ignition step is incorporated to remove organic matter.

The sample size will also relate to the method chosen. The presence of a heterogeneous population of particles dictates replication of analyses or analysis of large samples. The fusion of large samples presents many problems not the least of which is the need to purify from large volumes of salts.

It is also true that the degree of sensitivity desired suffers when very small samples are analyzed.

B. The degree of confidence associated with the analytical method should be higher than that defined for the mission. Although the statement may seem redundant, it is a point often overlooked. Sample should be subjected to the analytical procedure and the "degree of confidence" determined and related to that defined in the mission. Since the sample cannot be changed, the analytical procedure may require modification.

III. Sampling methods

A. Two methods for sampling soils for radioactive constituents have been used with success; these are the HASL method(2) based on the work of Alexander(3) and the NAEG method developed for the sampling of dry, sandy soils. The NAEG method lends itself more readily to true random and profile sampling; it does have the disadvantage of requiring more time per sample than does the HASL method. The mission will determine the method more nearly applicable.

The recently prepared draft guide AEC Regulatory 4.X is also suggested as a reference.

B. Important points related to the sampling of soils for radionuclide content are as follows:

1. As stated, the prime purpose is to obtain a representative and discrete sample hence classical methods, such as conservation auger, do not apply.
2. Cross contamination of samples must be avoided; cross contamination will bias final results, especially profile samples taken for the purpose of determining an inventory.

3. Volume and area of samples must be known precisely as factors in the calculation of overall aerial concentration.

IV. Choice of sample preparation method

Sample preparation is considered a part of sampling in this discussion; it relates to both the sampling method and the analytical method of choice. The cost of analysis (C2, page 3) is markedly effected by preliminary preparation; the purpose of the mission will dictate to some extent the sophistication of the preparation method employed. The best sample is the total sample, however the cost of preparation will dictate screening and/or aliquoting in many cases.

A heterogeneous particle size population will be present in certain types of emissions, e.g. fires or explosions.

Using the method of Leary(4) it has been calculated that a spherical particle of $^{239-240}PuO_2$, 44 microns in diameter, would assay at 86,000 d/m. In an actual analysis of one gram aliquots from the same sample, activities ranging from that level to 6 d/m/g were found.(5)
There is no known state of the art technique which will crush particles of those sizes to a uniform size and distribute them homogeneously throughout the soil matrix.

A prerequisite to sample preparation is the determination of a standard dry weight (105-110°C for 24 hours or to constant weight)(6) on a known vaolume related to a known area.

A. Preparation of the sample is dependent upon the analytical method of choice. The ideal aliquot to be analyzed will represent the entire sample, hence the aliquot taken should not contain stones or pebbles which will drop from the spatula and thus bias the sample. If stones and pebbles are present as such, the dissolution process will be extended and the cost increased. A pulverizing or flouring of the sample is most effective in reducing it to a form most amenable to analyses. However, in this respect samples with detectable activity must be processed in a dry box provided with an adequate air cleaning system; the treatment will produce fines (which have been shown to contain a high percentage of plutonium); the fines will be lost to the filters and the results biased.

A further point relates to cross contamination. A pulverizing mill used to prepare higher activity samples must be dismantled and decontaminated between samples; this is a costly procedure.

B. Definition of Excludable Material

The problems associated with A above can be solved in part by nested or contained mechanical sieving within an enclosure. With a single sieve, two fractions will be obtained---that passing, and that not passing. That passing will comprise the sample for analyses; that not passing is designated excludable material. Excludable in this sense connotes excluded from the primary "to be analyzed" sample. There remains to define the screen size and treatment of the "excludables".

A 10 mesh screen size for preliminary separation is acceptable. Root mat, large organic pieces, stones, pebbles, etc., which will not pass are removed from the sieve and weighed. Root mat and other organic detritus which is a definite part of the soil matrix (i.e., below soil surface level) should be added to the less than fraction and that fraction weighed.

The excludable rocks, stones, etc., may pose a problem; it has been shown that a very small percentage (less than 3%) of total activity is associated with this material(7) and hence may be discarded. However, a confirmation of "negligible percentage" may be desirable in which case the material may be acid washed and the washings added to the less than fraction or analyzed separately; if the washings are added, a second drying and weighing should be performed to obtain final weight of the "less than" fraction and to prepare it for following steps.

It is probable that some samples will contain above ground vegetation. Although this is part of the "collection system", it is not part of the soil system. The mission will define whether the total collector or the soil alone is of importance. In either case, it is recommended that vegetation be removed as a separate sample at ground level. Results of analyses can be weighted and combined if required.

C. The fraction passing 10 mesh is to be blended, mixed or ball milled; ball milling is recommended.

The NAEG procedure employs one (1) gallon paint cans protected by an outer brass sleeve. The protocol designates ball milling to the point where about 90% of the sample will pass a 100 mesh screen. The mission, cost, and analytical procedure will dictate the extent of ball milling. The sample is screened (100 mesh) and the weights of material passing and not passing are recorded. The less than 100 mesh material is the sample used for analysis. Drying at 110°C for 24 hours will generally brittle most material to the extent that it will ball mill properly, however, certain root material will not pulverize. The mission will determine the relative importance of root mat; if important, the organic detritus removed from the 10 mesh screen can be weighed, ignited, and the ash added to the sample being prepared for ball milling. The importance of base stem absorption (collection) has been pointed out by Russel. In certain missions, the effect of this collector should not be ignored.

It is well to maintain a library of prepared samples at least until the results have been compiled, reviewed, and accepted. However, recognize that a "second aliquot" analytical result may be disturbingly different from a former one. One then suspects a particle problem first (evidence to the contrary being absent) and the analytical method second.

D. Need for Special Treatment

A need for special problem or special study samples is not a unique occurrence and is noted here only to alert the reader that on-site decisions may be required to modify either the sampling or preparation procedure or both. The mission as well as physical conditions of the sampling point will determine factors which can be considered for modification. It is strongly urged that the boundaries set forth in Fig. 1 be used to guide the sampling.

V. Addendum

A. Controllable Variables

1. Analytical results are no better than the samples submitted; it is important to control all variables which can be controlled.

    a. The NAEG sampling protocol presents guidance to minify possible cross-contamination; the added effort required is minimal.

    b. Sample activity level can be controlled best by instrumental survey in the field to delineate those which are relatively "hot" or "cold" and thus alert the analyst to the aliquot size required for good statistics.

    A second approach involves a screening of samples in the laboratory by means of GeLi scan or other appropriate instrumental survey.

    The GeLi scan for

^{241}Am (60 keV) is especially helpful in the absence of fission products if ^{241}Pu forms a part of the radionuclide population; it is of little value in the presence of significant levels of fission products. Americium-241 x 10 has been used to indicate a possible plutonium concentration. However, the factor of 10 is variable and of questionable value in cases where plutonium concentration is low, hence the technique is limited to certain emission problems. However, the above can in certain conditions guide the analyst as to aliquot size.

c. Alternate Sampling Points - An initial statistical design for random sampling should consider the possibility that a sampling point cannot be used such as extended rock outcrop, hence some alternate random numbers should be available to be chosen should the need arise.

d. Soil Particle Size Definition - The size of soil particles which can be effectively sampled and meaningfully analyzed is limited. This item is closely related to "c." above. A sampling point consisting of one to two inch stones is of questionable value; a second set of random numbers should be available for such events if dictated by the mission.

2. Sampling Stratification

In the case of plutonium distribution (provided ^{241}Pu is present) a FIDLER instrument set to detect the ^{241}Am 60 keV energy peak is of value in delineating stratification and assigning a range of levels to be expected. Such an initial separation assists the analyst in grouping samples of like activity, reduces possibility of serious cross-contamination of samples in the field and guides the statistician in designing the sampling scheme. Other means of stratification may be used such as grass vs areas of brush or dry runs or creek beds vs upland.

3. Subsampling or Aliquot Size.

The aliquot size necessary to obtain some relatively constant level of activity per unit volume or per final plate for counting can be determined within reasonable limits by a GeLi scan as previously outlined. The activity per gram in many samples may be so low as to require a volume of soil too large to be accommodated by the analytical procedure and meet the suggestion of constant level of activity per plate. A relatively constant activity per unit volume or per plate simplifies counting (sample changing) and reduces gross cross-contamination possibility when lower level samples follow much higher level samples.

B. Certain Variables Require Further Study; In a Sense, These are Uncontrolled Variables at Present.

It has been pointed out that the distribution of particles relates to sample size. The distribution is one of size of particles as well as aerial distribution. In the cases of an emission or an inventory oriented mission, initial distribution or change in distribution with time are unknown. Investigation to define a representative sample under such conditions will be desirable.

The effect of soil structure and constituents on the representativeness of samples is unknown. Do particles per se transport through the soil profile; what is the chemical nature of the radionuclide in the soil constituents on transport, for example, the effect of organic matter on "solubility", chelation, or even insolubilizing of radionuclides? These questions are unanswered.

Such questions relate directly to profile sampling for inventory.

Are the analytical procedures equally effective with respect to all particles and their possible chemical states or forms? What sampling, preparation, and analytical methods apply to glassy material such as trinitite?

Are there conditions under which highly resistant forms (to chemical analyses) are produced -- and thus not accounted for? Tracer recoveries will not provide the answers.

Appendix B

Radionuclide Information

Ref.	Nuclide	Half Life (yrs)	Specific Activity (Ci/g)	Weight % of Nuclide in Weapons Mat.	Predominant α Particles (MeV)	(%)	X-Rays 10 - 20 keV				X-Rays 20 - 30 keV				Gamma Rays 20 - 30		keV		55 - 65 keV			
							keV	%	keV	%	keV	%	keV	%	keV	%	keV	%	keV	%	keV	%
(1)	Pu-236	2.85	520																			
(2)		2.85																				
(2)	Pu-238	86		0.04																		
(3)		86			5.50	71.9	13.6	4.60	17	4.31	20.2	0.58					43.5	0.04				
(4)		89.6	16.8		5.50	72			17	13							45	0.034				
(1)		86.4	17.4		5.50	72.2																
(5)			17.34					17-keV	band:	10.55												
(3)	Pu-239	24,000			5.16	72.0	13.6	1.74	17.0	1.63	20.2	0.22	band:	3.59	38.6	0.04	46.2	0.001	51.6	0.025	56.8	0.001
(4)		24,400	0.062	94.5	5.14	72.5			17	2.9			band:	2.9	39	0.003			53	0.007		
(1)		24,400			5.16	73.0									39	0.001						
(5)			0.0613					17-keV	band:	4.9			band:	4.9								
(2)		24,440		93.34			13.6	1.55	17.2	1.86	20.2	0.39	band:	3.8								
(4)	Pu-240	6,600	0.227	5	5.16	76			17	10							44	0.01				
(1)		6,540			5.16	75.5																
(5)			0.226					17-keV	band:	10												
(2)		6,580		6																		
(4)	Pu-241	13.2	113	0.5	4.9	0.004																
(1)		14.8																				
(5)			112.2																			
(2)		13.2		0.58																		
(3)	Am-241	433			5.57	85.3	14	16	17.4	15	20.8	2.0			26.3	3.1	43.4	0.01	59.5	38.4		
(4)		462			5.48	85			17	37					26	2.7	43	0.06	60	37		
(6)								17-keV	band:	37.6					26.4	2.5	43.4	0.07	59.6	35.9		
(1)		433															43.4	0.07	59.5	35.3		
(5)			3.22					17-keV	band:	37.6												
(2)		433			5.49		14	12	17.8	13	20.8	3	26.3	2.5					59.6	36		
(4)	Pu-242	380,000	0.00391		4.90	76																
(1)		387,000																				
(5)			0.0039																			
(2)		379,000		0.04																		

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1. Lederer(In press)
2. Wudnitz(1973)
3. Poston(1975)
4. Putzier(1966)
5. Tinney et. al.(1969)
6. Magnusson(1957)

APPENDIX C

TABLE C-1. FREQUENCY DISTRIBUTION TABLE FOR 80 ALIQUOT RESULTS ON REPLICATE SOIL SAMPLES FROM PENOYER VALLEY, NEVADA (from Bliss, 1973)

Interval	Midpoint	Frequency	Cumulative Frequency	Percent Cumulative Frequency
0. - 1.0	0.5	46	46	57.5
1.1 - 2.0	1.5	10	56	70
2.1 - 3.0	2.5	5	61	76.3
3.1 - 4.0	3.5	2	63	78.8
4.1 - 5.0	4.5	3	66	82.5
5.1 - 6.0	5.5	2	68	85
6.1 - 7.0	6.5	3	71	88.8
7.1 - 8.0	7.5	3	74	92.5
8.1 - 9.0	8.5	0	74	92.5
9.1 - 10.0	9.5	1	75	93.8
10.1 - 11.0	10.5	0	75	93.8
11.1 - 12.0	11.5	2	77	96.3
12.1 - 13.0	12.5	2	79	98.8
13.1 - 14.0	13.5	0	79	98.8
14.1 - 15.0	14.5	0	79	98.8
15.1 - 16.0	15.5	0	79	98.8
16.1 - 17.0	16.5	0	79	98.8
17.1 - 18.0	17.5	1	80	100.

Range of Data: 0.15 - 18.0 pCi/g

Interval Width: 1.0 pCi/g

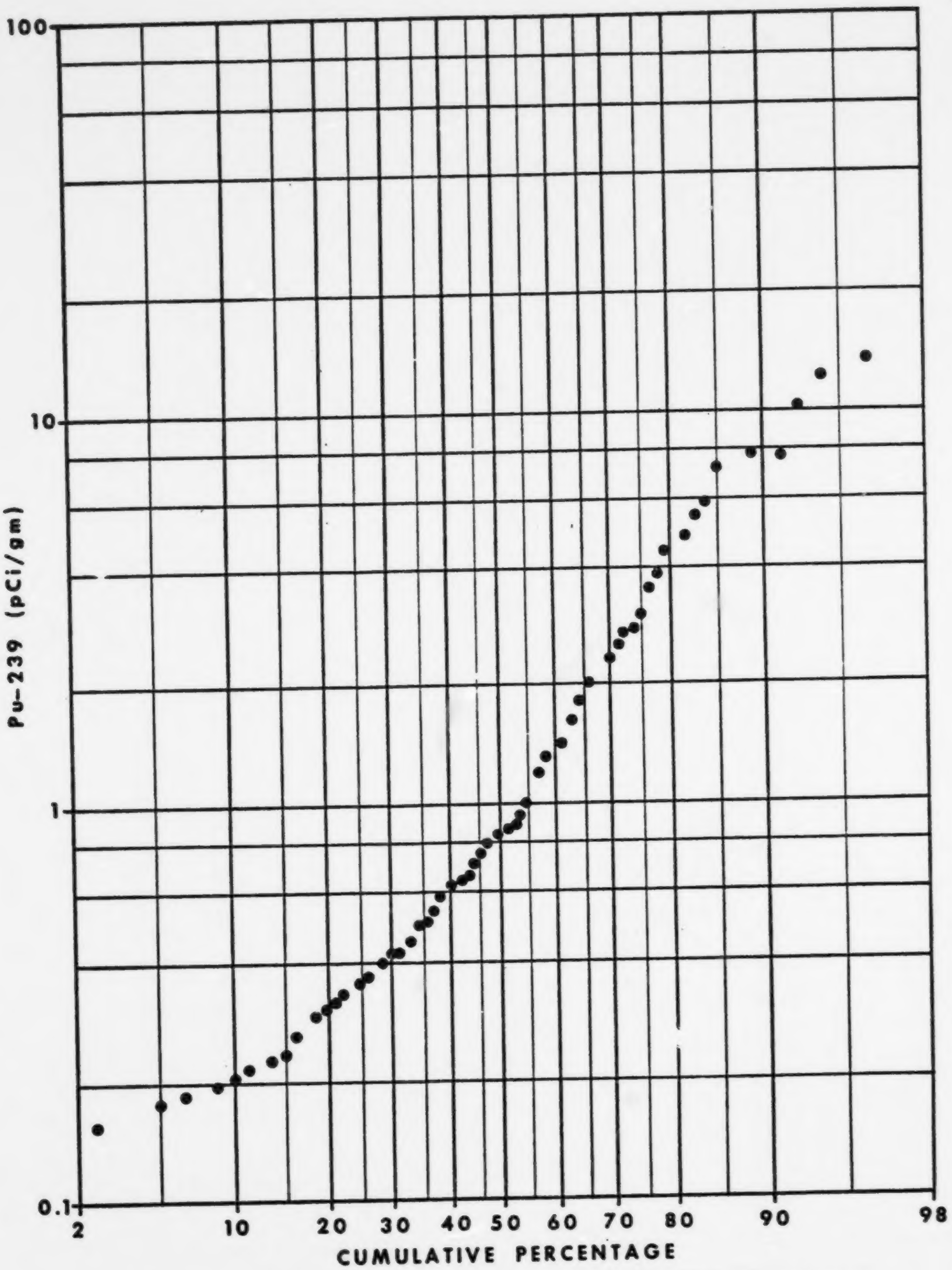

Figure C-1.· Probability plot of replicate samples, Penoyer Valley, Nevada.

TECHNOLOGY AND COSTS FOR CLEANING UP LAND CONTAMINATED WITH PLUTONIUM

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April 1978

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PREFACE

The Office of Radiation Programs (ORP) of the Environmental Protection Agency carries out a national program designed to evaluate public health impact from ionizing and nonionizing radiation, and to promote development of controls necessary to protect the public health to the environment. This report provides the technical information necessary for ORP to evaluate the environmental aspects concerning the costs and technology of cleaning up plutonium contaminated land. This document is part of the supporting documentation for the Federal Radiation Guidance for the cleanup of plutonium contaminated land areas.

David S. Smith
Director
Technology Assessment Division (AW-459)

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I. INTRODUCTION & CONCLUSIONS

A. Purpose

Plutonium contaminated* land areas can have an adverse impact on man if the plutonium is resuspended in the air (inhalation pathway) or if the plutonium is concentrated in plant life (direct ingestion by man or ingestion of animals that feed on the plants). Usually, the primary potential radiological hazard to man from land areas contaminated with plutonium results from the resuspension of the plutonium in the air as a particle. If the plutonium is plowed under, then the primary potential radiological hazard may be through ingestion of plant material. To reduce the potential radiological environmental impact on man, three general techniques normally can be utilized: removal of the plutonium from the land surface area, stablization of the plutonium on the land, or restriction of the use of the contaminated land. The purpose of this paper is to discuss the technological feasibility and the unit costs of employing these techniques or of utilizing several special techniques.

It is important to note that the Environmental Protection Agency is currently involved in developing environmental criteria for radioactive disposal and standards for the management of high-level radioactive waste. In addition, standards may be prepared for other forms of waste in the future. Many of the decisions regarding what must be done to meet these criteria and standards may not be made for several years. Currently guidance for cleaning up plutonium contaminated lands is reaching final development. However, the acceptable methods for cleaning up contaminated land and disposing the wastes have not been determined at this time. This determination will probably result from a cost-benefit analysis for each individual site. However there may be some types of disposal that will not be considered acceptable for waste removed from any contaminated site. In light of the uncertainty of acceptable waste disposal techniques for wastes removed from contaminated sites and facilities, this paper does not intend to select acceptable cleanup techniques nor compare the pros and cons of various cleanup techniques except on the basis of general costs. In this respect, there may be several techniques evaluated which may eventually be determined unacceptable as a result of future standards development.

With regards to the environmental criteria applicable to all radioactive waste, it has been recognized that there is great need to place a limit on the time period in which reliance can be placed on institutional controls. In applying this consideration to the subject of this paper, it is emphasized that reliance on institutional controls such as fences and guards cannot be found acceptable for long periods. The length of an acceptable period has not been determined at this writing but has been proposed as 100 years by EPA. Placing a time limit on institutional controls implies that the site or facility will evenutally revert to unrestricted use which, in turn, involves the many unresolved issues related to decommissioning. Thus, the use of institutional controls merely delays implementation of decommissioning standards and/or criteria. However, this delay can be quite effective for reducing environmental and public health risks in those cases involving shorter-lived radioactive materials.

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* Land areas are "contaminated" with plutonium if the resulting environmental impact of the area exceeds any Federal regulation or guidance.

The decision to apply any cleaning techniques to plutonium contaminated land areas should be based on Federal regulations and guidance and on a net improvement in the environment. The levels of radioactivity requiring clean-up or those to which the clean-up should proceed, as well as the aspect of control over the contaminated land areas are not addressed in this paper. The paper also does not address the clean-up of urban or industrial areas (our focus was on farmland, arid and semi-arid open land, prairies, mountainous areas, and forests) nor does it attempt to define or recommend which techniques could, or should be used, on the different kinds of land areas. Instead, the paper presents a discussion of the available techniques for plutonium clean-up projectssince each project site will vary relative to a large number of critical factors which significantly affect the costs and effectiveness of the various techniques. Some of these factors are physical in nature (such as the terrain, remoteness, climate, weather conditions, type of environment, use of the land, existence of vegetation, type of vegetation, type of soil). Other considerations are the restraints of surveying the area (which can cause delays) and the necessity for worker protection during clean-up (type clothing, respirators). These factors relate to the type and quantity of contamination. If there is any significant external radiation exposure to the workers, the costs and difficulties with performing effective clean-up operations may increase significantly.

B. Summarization

1. Technology. For any project, each of the general techniques - removal, stabilization, restricted use - will involve several or all of the following steps (or procedures): (a) radiological support, (b) stabilization, (c) removal, (d) packaging, (e) transportation, (f) ultimate ** disposal, and (g) restoration. The technology of and relationships of the techniques and the procedures are discussed in Section II. In summary, the procedures that may be employed with existing technology are:

(a) Radiological Support - A survey team employing such instruments as alpha survey instruments, and FIDLERS, or laboratory type instruments used in the field.

** The term "ultimate" means the resulting fate of the land at the conclusion of the specific technique. Other measures concerning the land may be required in the future and are discussed in the appropriate sections.

(b) Stabilization - Plowing, application of water, chemicals, vegetative cover, soil, sewage sludge, asphalt, or foam.

(c) Removal - Harvesting crops, removing vegetation and trees, scraping soil, vacuuming soil, flooding land.

(d) Packaging - Containerization of vegetation, soil, or solidified wastes.

(e) Transportation - Transport soil on or near the contaminated land area or transport containerized material to a waste burial ground or a Federal Repository.

(f) Ultimate* Disposal - restriction of land used; retention of the soil onsite (stabilization, concentration of soil as windrows, mounds, or in trenches); burial or storage at waste burial grounds, the Federal Repository.

(g) Restoration - application of soil, fertilizer, stabilizer, seeds, seedlings, shrubs.

2. Costs. There are several general techniques that can be utilized to reduce the environmental impact of an area contaminated with plutonium. A representative sample of the most often applied techniques is summarized in Table 4 along with their range of costs and average costs, and the advantages and disadvantages of each. A discussion of the costs is presented in Section III. Generally, the average✓costs (normalized to 1974 dollars) are:

(a)	Restriction of land use	$1100/acre
(b)	Stabilization	$2400/acre
(c)	Soil removal, onsite retention	$4800/acre
(d)	Soil removal, offsite disposal or storage at:	
	(1) a waste burial ground	$145,000/acre
	(2) the Federal Repository	$515,000/acre

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* The only truly "ultimate" disposal technique is elimination of plutonium contaminated soil by extraterrestrial means or transmutation. If any of the other disposal techniques are used, then it is always possible that additional measures may be taken in the future to restrict the land use or dispose of the contaminated soil.

II. TECHNOLOGY

The potential environmental impact of plutonium deposited on land areas may be reduced by a number of methods. Almost all of the methods can be categorized as one of three general techniques - removal of soil, stabilization of the land area, and restriction of the use of the land. In utilizing any one of these three techniques, several or all of the following seven procedures or steps may be necessary - radiological support, stabilization, removal, packaging, transportation, ultimate disposal, and restoration. The approach taken in this evaluation is to present (1) a general discussion of the three techniques; (2) a discussion of the seven procedures or steps; and (3) a discussion of their relationship with the three techniques. Also a discussion will be presented of several special techniques that are not generally applicable to projects involving the reduction of the radiological environmental impact of Pu contaminated on land areas. Finally, a short discussion of the effectiveness, advantages, and disadvantages of the techniques will be provided.

A. Removal

Removal includes options such as raking and grubbing out vegetation, stripping the top layer of soil by scraping, vacuuming, flooding, or applying a polyurethane foam cover (or other chemical cover) that removes the top layer of soil when the foam is removed.

Disposition of the contaminated soil and materials thus collected will depend on factors such as quantity of materials; contamination levels; local demography; meteorological and hydrological characteristics; land value; and land usage. Hauling will generally be required to dispose of contaminated soil offsite. The soil would have to be placed in containers or enclosed hauling vehicles prior to hauling. Local conditions permitting, however, burial may be preferable on or near the site (these methods are discussed under the stabilization section).

After an area is contaminated with plutonium, initially the bulk of the plutonium would be expected to reside very near the surface of the soil, on vegetation, on litter layer, etc. The distribution of plutonium also depends on the method of its release (airborne or liquid)

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* A description of the packaging requirements for plutonium contaminated soil are outside the scope of this paper. Current regulations for packaging are contained primarily in 10 CFR Part 71 and 49 CFR Parts 171 through 189.

and thus it may not always reside near the surface of the land. As weathering occurs and the area is disturbed by the forces of nature, part of this plutonium will be carried from the area, part will be distributed on the soil surface, and part will penetrate into the soil. The exact movement and distribution of the plutonium in the soil after weathering is very dependent upon climate, type of soil, and time elapsed before removal begins.

Raking and grubbing to remove vegetation and mulch will have variable effects depending on the time span since the plutonium was deposited on the land. As time passes, new vegetation replaces the old, new layers of thatch cover the old, and the plutonium is leached into the soil and removed from the land by weathering effects (such as wind or rain) so that removal of surface cover becomes less effective as a plutonium decontamination technique. The plutonium will also be distributed throughout the soil profile via root translocation and decomposition of the dead root material. This decomposition may also increase the uptake of plutonium by plants.

Removal of several inches of soil by scraping with graders, scrapers, or bulldozers is one of the most widely used methods of decontaminating an area. However, large industrial vacuum cleaners equipped with high efficiency particulate filters (HEPA) may be effective in decontaminating sandy soils or plowed land.[1] These vacuum cleaners have not actually been used to remove soil from land areas, thus their success can only be conjectured. Smaller vacuum cleaners may be useful devices where small areas are involved and the amount of loose surface material will not unduly load the equipment. Generally, a combination of the collection and removal techniques mentioned plus plowing the land will provide optimum results (see the discussion in the stabilization section).

Polyurethane foam, which is applied by spraying, has been used successfully on small areas.[2] After setting, it is sectioned and removed. The top layer of soil, along with small rocks and detritus, adhere to the underside of the foam and are removed with the foam. Since this technique is experimental and also very costly, it would probably not be used extensively as a soil removal technique at this time.

B. Stabilization

Stabilization includes techniques such as plowing the land to cover the top soil layer, scraping the soil into trenches, contouring the land in windrows or mounds (this is also a removal technique), covering the land with soil or vegetation, and applying chemical stabilizers to the land area. It should be noted that usually stabilization is only a short term solution which may often require a more permanent clean-up technique in the future. Plowing can be used to invert the contaminated top soil to place it beneath a cover of subsoil; thus, providing a covering to prevent resuspension and also to achieve dilution. It has the disadvantage, however, that subsequent removal operations, should they be required, would be rendered much more difficult because of rearrangement and dilution. Normally the dilution afforded by plowing would place the area in unrestricted use. Thus, future clean-up efforts or surveillance of the area would not be necessary.

Scraping the land into windrows or mounds has been used to minimize the dispersion of radioactive contamination by wind action. Windrowing could be accomplished by first scraping (blading) the top surface of the soil and detritus into parallel rows. Soil below the top layer is then bladed over the contaminated material to form windrows. Mounds are formed by pushing the soil into large piles. Then, a clean earth cover or chemical stabilizer can be applied to fix the plutonium. Establishment of a vetetative cover, controlling access to the mounds, and using other limitations on land use can be employed to maintain the integrity of the windrows or mounds. The long term integrity of the windrows and mounds are very dependent on the effectiveness of the stabilizers to prevent erosion of soil or resuspension of plutonium while the vegetative cover gains a foothold. If the stabilizers are not effective, then the expense of an alternative technique will also be incurred. The soil can also be scraped and deposited in trenches. The trenches can then be covered with uncontaminated soil by backfilling. The problems associated with this technique are similar to the problems with windrows or mounds except the contaminated soil will not be as susceptible to the wind and rain erosion.

C. Restriction of Land Use

Restriction of the contaminated land can often be accomplished by placing a chain link fence (with or without barbed wire at the top), signs to warn people, and a temporary guard service (if the area is reasonably sized). It should be noted that restriction of land use is only a temporary solution. Thus, either additional maintenance costs or a more permanent clean-up technique may be required in the future. This technique by itself would not prevent resuspension of the plutonium but would be useful to prevent access by people and animals. The initial cost of the fence is not prohibitive; however, a 24 hour per day guard service plus the maintenance of the fencing will be a continuing expense that can be expensive if maintained for many years. If fencing is continued for a long period of time, the land is relegated to a non-productive status. Fencing may be effective if the activity levels of the contaminated area are not very high and the land can not be used for any worthwhile purpose (e.g., remote areas where land is either not fertile or not easily accessible). It should be realized that other decontamination techniques may have to be utilized in the future if permanent fencing is either not feasible, ineffective, or impractical. It should also be stressed that there may be an adverse psychological impact upon the population of scattering fenced areas around the countryside, perhaps with armed guards.

Restriction of the use of some types of contaminated land can be done by changing the use of the land. For example, farm land, commercial forests, or any type of land can be bought by the Federal Government and relegated to a non-productive status; national parks can be closed; land can be rezoned; and crops can be purchased by the Government. The costs of farmland, tree forests, and crops are discussed in Section III-D under Economic Losses. However, an exhaustive analysis of this technique is not included in this paper.

D. Special Techniques

Several techniques that may not be utilized very often for varying reasons are discussed in Appendix B, Part 1. They are stabilization by application of asphalt or sewage sludge, decontaminating snow covered areas, flooding as a soil removal technique, and taking other preventive type measures.

E. Procedures for a Project

The seven procedures (indicated in the first paragraph of Section II) are discussed in detail in Appendix A, Part 1. Table 1 presents the relationship of these seven procedures (first column) to stabilization (decontamination techniques I) and removal (decontamination techniques II and III). The stabilization techniques that are indicated in Table 1 are plowing, application of chemical or vegetative stabilizer, and application of a soil cover of two different depths. The only procedures that are utilized for stabilization techniques are radiological surveillance, stabilization, vegetative removal (where appropriate), fencing (optional), a guard service (optional), and restoration (mainly for erosion control).

The removal techniques that are indicated are scraping soil into windrows, mounds, or trenches, vacuuming soil into trenches, and vacuuming or scraping soil with offsite disposal at waste burial grounds or the Federal Repository. Removal with onsite disposal of the soil (decontamination technique III on Table 1) includes all the procedures.

Restricted use of the land (not included in Table 1) would involve radiological surveillance, stabilization (optional), vegetative removal (where appropriate), fencing, and a guard service (optional). A discussion of restricted use of the land by fencing is presented in Appendix A, Section F, Part 1.

III. Costs

The costs presented throughout this report, unless indicated otherwise, are normalized to 1974 costs.[12,13] The regional variances of labor and material costs for construction jobs in large cities in the United States are presented in Table 3. The average costs, and their ranges, presented for the various techniques and procedures should be considered as general guides since the costs for a specific project are dependent on a variety of parameters such as weather conditions, terrain, soil conditions, accessibility of the site, age of the Pu, moisture in the area, type of vegetation, use of the land, and many other parameters. Because the cost ranges are often very large, average costs may not be very useful for a particular project. Also for unusual projects the costs may even be outside the cost ranges presented.

A. Removal, Stabilization, and Restriction of Land Use

The average costs, and the range of costs, for the removal and stabilization techniques presented in Table 1 are presented in Table 2. The bases for these costs for each technique, and the procedures listed under each technique, are presented in Appendix A, Part 2. The sections in Appendix A, Part 2, are designated in the same manner as the sections in Table 2. The costs for restricting the use of land include radiological surveillance costs (See Appendix A, Part 1, Section A), stabilization (optional costs not included in this evaluation), and ultimate disposal costs being fencing and a guard service (optional costs not included in this evaluation).

On particular projects, the radiological surveillance costs will vary depending on the time required to complete a decontamination project and the activity levels of the contaminated land. The stabilization techniques and stabilizers are dependent primarily on the extent of plutonium resuspension, availability of stabilizers, and the type of soils. Vegetation removal techniques, or course, are primarily dependent on the type and extent of vegetative growth, the activity levels on the vegetation, and the clean-up techniques to be employed. The requirements to package, transport, and dispose of the soil at a waste burial ground or Federal Repository are dependent on the activity levels of the contaminated soil that will be removed from the site for offsite disposal. The primary parameters that affect restoration techniques are the aridity of the area and the type of vegetation that will be effective to prevent erosion and also adequately revegetate the land. Soil removal costs are dependent on a number of factors, such as land terrain, soil moisture and characteristics, site accessibility, removal equipment availability, weather conditions, depth and distribution of plutonium in the soil, activity levels, equipment operating efficiency, operator techniques, distribution and size of rocks.

It appears that the soil type (e.g., sandy, clayey, tilled land, etc.) determines the selection of the soil removal technique, and the resultant soil removal costs, more than any parameter, except plutonium activity levels. The low end of the cost-range for removal would apply to sandy type soils and the high end of the range would apply to hard, clayey soils.

The total average costs and their ranges for the techniques previously mentioned are presented in Table 4. Except for the costs for restricting the use of the land, the costs presented are extracted from the TOTAL PROJECT COSTS presented at the bottom of Table 2. For restricting the use of the land, the costs are two fold - the surveillance costs extracted from procedure or step 2 of Table 2 (average $600/acre and range $250 - $1100/acre) and the fencing costs extracted from Appendix A, Part 1, Section F (average costs $500/acre for a 100 acre site and range $100 - $2000/acre for the low range of a 1000 acre site to the high range of a 10 acre site).

In summary, the average costs of each technique are:

1.	Restriction of land use	$1100/acre*
2.	Stabilization (Techniques 1 and 2 of Table 2)	$2400/acre*
3.	Removal with onsite retention (Techniques 4A, 4B, 4C, and 5 of Table 2)	$4800/acre
4.	Removal with offsite disposal or storage	
	(a) Waste burial ground	$145,000/acre
	(b) Federal repository	$515,000/acre

B. Special Techniques

The special techniques considered are application of asphalt or sewage sludge as a stabilizer, decontamination of snow covered land, flooding as a soil removal technique, and removing soil under a protective air shelter. A discussion of the costs of these techniques is presented in Appendix B, Part 2. Also, a discussion of costs is presented for a 24-hour guard service (Appendix A, Part 1, Section F), disposal of solid waste in the ocean (Appendix A, Part 1, Section F), and application of polyurethane foam as a soil removal technique (Appendix A, Part 1, Section B).

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* These costs do not include on-going maintenance costs. A discussion of these costs is presented in other sections of this report.

C. Plutonium Clean-up Experience

The available cost data for actual plutonium decontamination projects are presented as total project costs except for the proposed Enewetak Islands project which ran to $70,000/acre.* The costs for four of the projects ranged between $9,000 to $83,000 per acre for a wide variety of projects, while three of the four projects ranged between $9,000 to $26,000 per acre. The Niagra Falls clean-up was not plutonium related. The fourth project, the decontamination at Palomares, Spain, included many extra costs and high transportation costs that probably would not be included if similar decontamination were performed in the United States. The fifth project (at Rocky Flats) involved manual removal of soil under a small portable metal enclosure; thus, the project was time consuming and costly. However, costs were not available for this project.

The costs for the four projects are presented in Table 5. They have not been normalized to any particular year, but they are still an adequate presentation of general cost ranges for decontamination projects. It should be realized that each of these situations was unique, hardly representative of average land clean-up conditions and should not be used as a basis for deriving average cost estimates.

D. Economic Losses Resulting from Pu Decontamination Projects

The loss of crops or use of the land as a result of a decontamination project should also be considered in an economic analysis of Pu clean-up efforts. Tables 7 and 8 present the potential economic losses for farm crops during 1973 and for forest trees in the national forests during 1974. The market value of farmland in the United States for 1974 averages about $310 per acre with a range of $65 - $2100 per acre as an average value by state (48 states).[54] In summary, the average losses would be about:

crops	$170/acre/year
forests (commercial)	$2650/acre/year
farmland	$310/acre

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* See the references for details of the costs. The costs for Enewetak are estimates since this is a future project. Costs are per cubic yard (c.y.), assumed 538 c.y./acre of soil removed (4 inch depth). The actual costs are estimated to be about $70,000 per acre for a 4 inch soil removal. A 4 inch removal is assumed throughout this report for consistency. If a different soil depth is chosen, the cost can be estimated directly. The Army Corps of Engineers indicated that these costs are about a factor of 2.8 higher than the costs for decontaminating the same land in the United States.

APPENDIX A. Bases for Costs in Table 2

PART 1. General Information: Section A-H*

Section A. Radiological Support**

The radiological surveillance costs are based on the assumptions that a surveillance team consists of one health physicist (GS-13), five monitors (technicians GS-7), and one electronics maintenance man.[14] One team can adequately utilize five alpha survey instruments and five FIDLERS (Field Instrument for the Detection of Low Energy Radiation) during a project. A work week consists of 12 hours/day, 7 days/week. The team would be at the project a short time prior to decontamination procedures, the entire time during the decontamination, and a short time after the decontamination is completed. The costs of a survey team also include the travel and per diem expenses and laboratory costs for analyzing samples. The following indicates each of these costs for a 100-acre plot:

One Health Physicist @ 84 hours/week $868/week
Five Technicians @ 84 hours/week $536/week (each man)
One Electronics Maintenance Man @ $10/hour @ 84 hours/week $1060/week
Per Diem for Seven workers @ $25/day/worker, $1,225/week
Five FIDLERS @ $3,000 each, ten-year lifetime $1,500/year
Five Alpha survey instruments @ $1,200 each for 10 years $600/year
Travel, one way .15/mile
Laboratory analysis, $100/sample @ 1 sample/acre (radiochemistry analysis in field)
For a 1,000 mile trip, the costs per acre per week are calculated to be $37/week/acre plus $300 travel for the project plus $100/acre for sample analyses.

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* Appendix A, Part 1, discusses the general procedures and costs for the seven general techniques that are applicable to any decontamination project. The techniques referred to in Part 1 are presented in Tables 1 and 2.

** Radiological support costs were based on the assumption that the survey team consisted of individuals and equipment from EPA's ORP Las Vegas Facility. If the survey team does not come from EPA, then the costs could be quite different from those presented.

It is estimated that projects ranging from one month to six months will cost: $250 - $1100/acre with an average of $500/acre for a three month project. These costs are indicated in Section A.1 through A.6B of Table 2, since radiological surveillance is common to all plutonium decontamination projects of the same time period.

Section B. Stabilization

In most cases, experience with stabilization of contaminated land areas has been confined to "test" sites and small research areas. As a result, many of the stabilization techniques that have been considered are experimental. The success of specific stabilizers may well vary where the concentration and source of contamination vary, or where the land surface or climatic characteristics are dissimilar. In addition, since the same techniques have not been tested on all surfaces, it is difficult to accurately assess the advantages or disadvantages of any particular method since general applicability has yet to be established. It is possible that alternative methods or types of stabilization would prove to be more successful for decontamination purposes.

The experimental nature of previous decontamination situations also offers another important point for consideration. In most of the experiments, the goal was to determine the effectiveness or benefit of a particular technique or stabilizer -- and not to determine costs. As a result, in almost all cases, no records of cost had been maintained. Inability to acquire accurate cost figures based on actual decontamination experience necessitated development of cost figures based largely on assumptions. Thus, it was not possible to verify these cost estimates or the basic assumptions through comparison with empirical experience.

The types of stabilization methods experimented with thus far fall into five categories. These categories and their costs are presented in Table 6. The first type of stabilizer is only temporary, e.g., application of water. It is likely that water would be used following a contamination incident to minimize the resuspension of plutonium particles until the seriousness of the event could be thoroughly evaluated and a clean-up plan initiated. In situations where expediency is advantageous in reducing potential exposures, water offers the desirable characteristics of being readily accessible in most areas and thus enabling timely action to be taken. At the same time, it does not represent any irreversible commitments as far as other clean-up operations are concerned. Water can be applied to any type of soil with a similar degree of success in preventing resuspension. The cost to apply water for a 0.3 inch depth[4] will run about $220 per acre.[16,42]

There are several ways water can be used and the amount of water required depends on the effect desired. Distributing a fine mist into the air several feet above the contaminated surface until the surface is wet to about a depth of three tenths of an inch has proven to be effective in holding down dust for an hour. (This effective time will vary locally in response to differing climatic conditions and evaporation rates). The efficiency for preventing resuspension is about 50%, ranging from 20-90%.[4,17] Direct wetting by hose also has a similar effective life, though the pressure of the water in this case causes a mixing in the upper centimeter of soil and thus would cause a somewhat more effective fixation of the contamination.[15] The obvious problem here is that unless other measures are taken almost immediately, it becomes necessary to rewet the contaminated area hourly or risk resuspension of plutonium activity.

Water can also be used to flood a contaminated area. At least one inch of water may be applied (with care taken to divert any runoff) to enhance natural weathering and leach the contaminant into the soil, thus reducing the amount of contaminant available for resuspension by almost 85%.[4,17] This use, while still only "temporary" does not require the frequent reapplication of the wetting methods and still is fairly inexpensive. It would be possible to effectively apply the water spray to almost any unvegetated surface without being restricted by the local terrain. Flooding on the other hand would be more limited by the terrain since an essentially flat surface would be necessary in order .to maintain standing water on the contaminated area. It might even be necessary to construct low dikes around the perimeter of the area to help contain the water.

There are several problems created for later clean-up efforts when water is applied in the initial stages of decontamination. For example, when the polyurethane foam is applied to a water saturated surface, a component in the foam reacts with the water and tends to form a void between the foam and the surface of the soil.[2] A contaminated surface treated in this manner would not be stabilized securely; thus, pick-up of the contaminated soil by the foam would be erratic. This situation could be modified by allowing the soil to dry to a just-damp state before application of the foam.

The amount of moisture left in the soil would also impact on the success of several of the other stabilizers, although not as significantly. Since most of the chemical stabilizers are mixed with water when they are applied, a limited amount of excess moisture in the soil can be compensated for by increasing the concentration of the chemicals during application.[18] An increase in concentration is not effective, however, on saturated soils.

Both the asphalt-like surfaces and soil can be applied over moist earth. In these cases, heavy machinery is involved in applying the surfaces and such machinery does not maneuver well in mud. Again it would be necessary to allow the soil to dry out somewhat before continuing with decontamination.

Flooding could complicate removal procedures (though not seriously) if the flooding were repeated over an extended period of time. Each subsequent flooding would leach the contaminant deeper into the soil, possibly dispersing it deep enough to necessitate additional passes of the removal equipment. Neither this problem of leaching nor the problem of poor adherence due to high moisture content are insurmountable.

A second type of stabilization that has been experimented with seals the contaminant more permanently by covering the surface with a layer of an impermeable substance, such as fast-cure road oil, or asphalt emulsion. In both cases, there is only limited information available on their use as stabilizers.[4] Both substances are reasonably easy to acquire and both can be applied with conventional road-oil or asphalt spray trucks.

At the Nevada Test Site, road oil was applied to soil to a thickness of from one-half inch to two inches. The oil formed a crust which maintained its integrity (while exposed to desert weathering) for almost 5 years and would have required an additional thin coating after that time for maintenance.[19] The tests proved this technique as somewhat successful in a desert climate but there are two major drawbacks with the use of rapid cure oil: its surface remains tacky for quite some time after curing, and depending on the locale, it may be stocked on a seasonal basis only, thus, it may not be readily available at all times.

The fact that the surface remains tacky could cause problems for animals particularly if they were to wander inadvertently into the area. Although experience has indicated that animals tend to avoid such surfaces, additional research should be conducted to see just what effect an application of road oil would have on localized animal life. The total cost of the road oil and its application would be about $660 per acre.[20]

There is even less information available on the use of asphalt, but its use has been considered as a means of containing plutonium on a temporary basis until contamination can be removed, or on a permanent basis (up to 10 years).[20] (This is discussed in the section on Removal). In the first case, about 1/16-inch of asphaltic emulsion could be spread at a cost of about $1,100 per acre.[21] This treatment locks on the surface and makes removal of the contaminated soil more efficient. In the second case, the asphalt surface would be expected to have a longer life and would be at least 1/2-inch thick. At one dollar per square yard[20] for material and application costs, it would cost about $4,800 per acre to pour the asphalt. In order to weatherproof the surface, a sealer would have to be applied at a cost of $0.05 per square yard[22] bringing the total cost to about $5,100 per acre.

Any area stabilized with asphalt would also require that adequate drainage be provided. Since the finished surface is impenetrable to water, any rain would tend to accumulate in low spots and then wash over the surface in sheets. Gathering volume and momentum at the edge of the asphalt, the flow could erode peripheral soil and undercut the prepared surface, threatening its integrity. Prepared drainage would minimize this concern and also allow for control and possible diversion of run-off in the event that it became contaminated. The asphalt surface would also be subject to the "road edge effect." As the surface would crack with time, moisture would collect in the cracks where plants could germinate and grow. With the maximization of plant growth, the surface would be further destroyed, threatening the possibility of recontamination.

The road oil and the asphalt could be applied over small amounts of certain vegetation and both prevent resuspension of plutonium over fairly long periods of time. However, they are unsightly, and the areas stabilized in such a manner will require constant surveillance since the plutonium would be fixed underneath. Also, the surfaces covered would remain useless while stabilized -- unattractive to both plant and animal life.

The third type of stabilizers are those loosely described by the term "chemical binders." (Most of the tests that have been conducted with such substances were conducted with specific brand name binders and although these brand names are merely representative of the field, it simplifies the following discussion by utilizing the brand names).

Chemical binders are typically substances that were developed commercially to be applied, along with seed and mulching material, on bare soil or slopes to bind the surface and assist in erosion control. They are diluted with water to the appropriate strength and applied in a spray form by means of a hydroseeder or a tank truck equipped with a spray bar.[23] In their normal use, they would secure the soil surface and the seed in place until the seeds can germinate and provide the vegetative cover to prevent erosion by wind or water. The use of such chemicals has become increasingly popular with construction companies; thus, they are not difficult to procure. The time period after an accident that these chemicals could be procurred is highly dependent on the area of the country (remoteness, proximity to a company's distribution network, etc.); however, they should be available at the site in no more than a few days.

Because of the stabilization properties of these binders, they have been considered for use in preventing resuspension of plutonium. The chemicals can be applied without seed or mulching materials and at different strengths to produce surfaces whose integrity vary with time. The less concentrated application could be ideal for temporarily preventing resuspension, while a stronger concentration may provide adequate protection for up to five years.

The following are binders which have been tested successfully as stabilizers for land contaminated by radionuclides other than plutonium.

(1) Geo-tech: A clear non-toxic resin which when applied penetrates about 1/4-inch into the soil. It binds the soil against degradation by wind and water yet also allows moisture to pass through. It will maintain its integrity even after exposure to foot traffic, but begins to deteriorate after about one year if exposed to the sun. No tests have been conducted beyond that length of time but some sources feel it would only be necessary to reapply the binder every 5-10 years to prevent resuspension of plutonium since this product is very effective in controlling dust. The cost for materials and application of this stabilization material would run about $520 per acre.[4,24]

(2) Norlig and DCA-70 were experimentally selected over similar products to stabilize the Tuba City mine tailings. They penetrated 3/4-inch and 2 inches respectively and became water insoluble after emplacement. No time period was available for the integrity of these crusts (the initial work was completed in 1969). The costs of these applications per acre are about $240 and $440 respectively.[25]

Other "binders" that could possibly provide useful stabilization in plutonium decontamination efforts include:

(1) Dust Control Oil: Sales representatives tout a 90-95% minimum efficiency of this product in reducing dust and preventing resuspension of a contaminant.[21] Several references indicate the range to be 70-90% with an average of about 85%.[4,17] It does not form a hard surface, and it can be plowed and revegetated after 3 or 4 years. Depending upon the strength of the solution applied, dust control oil can cost between $360 and $730 per acre to apply.[21]

(2) Petroset: A petroleum resin in a rubber base which penetrates to a depth of 1/2-inch and will bind the soil for at least 9 months. Like the Geo-tech, it allows moisture to penetrate and can be applied to either encourage or discourage revegetative growth. Cost for application to an acre is about $1,200.[16]

(3) Polyvinyl Acetate: Sprayed with conventional equipment, it penetrates 1/4-inch into the soil surface. This treatment protects the surface from wind and water erosion and will last a year with some minor deterioration from sunlight.[18] Depending on the concentration of the material that is applied, costs vary from $200 - $700 per acre.[26]

These chemical binders only represent a sample of many specific brands and types commercially available. It appears that there are many such chemicals which have properties applicable to the plutonium clean-up problem - but further laboratory studies need to be conducted in order to determine which would be most effective for this application.

The fourth type of stabilizer is unique in that it was tested as a stabilizer but was also found to provide excellent scavenging characteristics for contaminated soil. The stabilizer used was polyurethane foam applied to test plots at Rocky Flats at a thickness of about 2 inches. The foam encapsulated the rocks and debris on the surface and when removed, 85% of the contaminated soil was also removed.[2]

The foam is a special case -- it adheres to a variety to substrates; it can be used readily in a wide range of weather and topographic conditions; it resists environmental decay for periods up to 2 years,[27] and it can pick up stones up to three inches in diameter. Polyurethane is used as insulation in houses and other buildings so it is available, though perhaps in limited quantities, in most areas of the country.

Perhaps the major drawback about the foam is that its costs are much higher than some of the other stabilizers. Of course, it was not intended to be used as a stabilizer as the others are, but a price quoted from a local polyurethane dealer puts the cost of the required 2-inch thick covering of polyurethane at 65¢ a square foot or approximately $28,000 per acre.[22] This cost is prohibitive over areas as large as an acre -- and estimates run that it would take 2 men at least seven and perhaps as long as 12 days to apply it to a one-acre land area.

A fifth type of stabilization involves the use of soil and/or vegetative covering. This method would leave the contaminated area in a generally more attractive and usable condition than any of the others, and it would also serve as a restoration process. However, covering an area with soil does not guarantee vegetative cover, especially in arid regions. Generally, a soil cover of at least 4 inches would be applied over the contaminated surface. This would limit resuspension and would also provide a surface which could be readily revegetated, as desired, once amendments (like lime) were applied. (Some amendments appear to be capable of minimizing the uptake of plutonium by plants).[6] In most areas, the revegetation process could be enhanced by the controlled application of water to encourage plant growth. Surface occupancy would be allowed only if continuing surveillance determined that the underlying contamination was not brought to the surface by man-made or natural disturbances.

The cost of soil cover is rather high, with loam running between $1,500 and $3,300 per acre for a layer 4 inches in thickness, and topsoil running from $3,300 to $5,700 per acre for the same depth.[28,29] Increasing the thickness of the layer increases protection and reduces the risk of resuspension, but is also proportionately more expensive.

In most cases some type of stabilizer will probably be applied to a land surface after a plutonium-contaminating incident requiring remedial actions. The "fixing" qualities of stabilizers allow the necessary control over resuspension of the plutonium and provides time to determine an appropriate clean-up plan. The stabilizer selected will be determined by the nature of the incident and the specific decontaminatinn benefits desired.

Section C. Removal

Costs were estimated for two types of removal: (1) vegetation or crop removal and (2) soil removal. For all the techniques analyzed, vegetation removal would be optional depending on the type of area decontaminated, type of vegetative growth, concentration of contamination, and extent of clean-up required. The plutonium removal efficiencies vary considerably from 70% for raking mulch to about 30% for mowing or harvesting.[3] Vegetation removal costs were calculated for three general categories: (1) clearing and grubbing non-wooded areas, (2) clearing and grubbing slightly wooded areas, and (3) removing trees. Note that clearing and grubbing for non-wooded areas includes mowing grasses, raking mulch, flailing crops, harvesting crops, or using a combine. The costs for these methods are generally at the low range of the costs that are presented in Table 2. The costs include overhead and profit of about 25% and hauling the debris offsite at about a 20-30% incremental cost. There are no costs included for either incinerating the trees or for special packaging of the vegetation or trees which are to be taken to a solid waste burial ground or Federal respository. There has been no experience with such contamination to indicate what type of disposal would be appropriate. If the trees are to be incinerated, it will be necessary to use special particulate filters to prevent resuspension of plutonium. No data was available on this procedure. These costs, on a per cubic yard basis, are expected to be comparable to costs for packaging and transporting contaminated soil. The costs for each type of vegetation removal are assumed to be:

Technique	Costs	References
clearing and grubbing non-wooded areas	$ 500/acre	30, 31, 32, 33, 34
clearing and grubbing slightly wooded areas	$1,250/acre	32, 33
removing trees	$2,000/acre	33, 35, 36

The ranges of data are large, as shown in Table 2; however, the assumed costs are reasonable to categorize the costs since many factors contribute to the costs of a specific project. Some of these factors are type of undergrowth, size of trees and thickness of forrested area, terrain, capability of using large machinery, contamination levels of land and vegetation, and ultimate disposal of vegetation.

Soil removal may be accomplished by using scrapers (towed types, bulldozers, carryall scrapers, self-loading grader, etc.), industrial sized vacuum cleaners (on plowed land or sandy soil), and polyurethane foam. Scraping involves either (1) scraping the soil into windrows or mounds, (2) scraping the land and carrying or pushing the soil to trenches for burial, or (3) scraping the land and picking the soil up for packaging and offsite disposal. Industrial vacuum cleaners can be used in conjunction with HEPA filters to remove the soil after it has been plowed or scraped into windrows or mounds and have it either transported to a trench for disposal or to be packaged for offsite disposal. The removal efficiencies for scraping range between 60-100% with about a 90% average. Scraping in combination with plowing and/or backfilling have combined Pu removal efficiencies of about 98%.[33,37] Polyurethane foam may be applied to some types of soil as a stabilizer (as discussed previously). The soil particles are bound to the foam and the foam can then be removed by a front end loader, incinerated in a special Pu incinerator, and the contaminated ashes packaged for offsite disposal.

The ranges of costs for scraping land can be below \$100/acre to as high as several thousand dollars per acre depending on many environmental factors including moisture of the soil, weather, temperature, terrain, type of equipment, availability of equipment, access to the land, type of soil, contamination levels, number of scraping passes necessary to remove contamination, number of rocks on the land, etc. The assumed costs are given for calculational purposes only but the general costs presented should be adequate to compare the different techniques. The costs for just scraping 4 inches of soil are around \$200 - \$600/acre,[3,4,16,33,34,37-40] while the costs for scraping, digging, and backfilling a trench are about \$1,700/acre.[32-35,41,42] Using an industrial vacuum cleaner to remove soil followed by disposal in a trench costs around \$2,450/acre.[2,32-35,41,42] Scraping plus packaging the soil in containers as a preparation for transportation and disposal offsite averages around \$800 - \$1,000/acre, with the overall range of costs of about \$450 - \$2,800/acre.[1,3,4,16,33-35,37-42] Application and removal of polyurethane foam costs about \$28,000/acre.[28]

Section D. Packaging

The plutonium contaminated soil that must be transported to a waste burial ground or the Federal repository for ultimate disposal must be packaged in lined containers (usually 55-gallon drums), loaded and transported by rail and/or truck to a licensed burial ground. The major items that enter into the overall packaging costs include the cost of drums, freight charges, return of the trucks (or rail car) to the project site, labor for loading and unloading, truck driver costs, and capitalization.

The costs for packaging wastes for disposal at waste burial grounds and the Federal repository ranged between $6.80/cubic foot to $10.20/cubic foot including transportation costs for a nominal 1200 mile round trip.[43] The costs from reference 45 were for a 600 mile round trip, but would still be in the same range for a 1200 mile round trip. For a nominal removal of 4 inches of soil from an acre of land, the cost becomes about $100,000 to $150,000 per acre. For calculational purposes, it is assumed that the packaging costs for waste sent to a waste burial facility is $100,000/acre and that the packaging costs for waste sent to the Federal repository is $150,000/acre. For transportation distances significantly larger than about 1,200 miles round trip, the added costs are presented in Apeendix A, Part 1, Section E, under Transportation.

Section E. Transportation

Transportation includes two types of mobilization: (1) moving soil by a scraper or dump truck several thousand feet to form mounds of soil or to a trench for burial and (2) transportation of containerized wastes to the licensed waste burial grounds or the Federal repository.

(1) Moving soil by a scraper - Building mounds or transporting soil to trenches requires more time and effort than scraping the soil into windrows. For estimating the costs of a job requiring a haul, the costs discussed under Removal, Section C of Appendix A, Part 1, include the costs for a 1,000-foot haul.[32-34] The costs for longer hauls were extracted from this removal data for distances of 300 feet to 5,000 feet. The range of data is greater than an order of magnitude, mainly because it appears that the type of scraper and the actual haul distance are very critical. (Large scrapers add smaller costs for longer hauls and extra costs for the last few 1,000 feet is less than the first few thousand feet). All the costs per acre were estimated assuming a removal of 4 inches of soil, or 538 cubic yards, per acre, The range of costs for a 1,000 foot haul are $80-$880/acre with an average of about $300/acre.

(2) Transportation of containerized wastes - The costs for packaging (Section D of Appendix A, Part 1) include a nominal transportation distance of 1,200 miles. The extra transportation costs would be included only if the distance were greater than 1,200 miles. The costs range from \$0.05/cubic yard/mile to \$0.20/cubic yard/mile.[32-34] For a removal of 4 inches of soil, or 538 cubic yards, per acre, the range is about \$27/acre/mile to \$108/acre/mile. The assumed value is \$0.10/cubic yard/mile or about \$54/acre/mile. For each 100 miles, the extra transportation costs would be \$5,400/acre.

Section F. Ultimate Disposal

A plutonium contaminated area may be restricted from public access by placing a fence around the area and establishing a 24-hour guard service at the area for a short-term (a few years). A guard service, however, is usually not necessary and is not a normal requirement. Prior to fencing, the contaminated material may have been stabilized by plowing, covered by vegetation, chemical stabilizer or soil; or buried in a trench. Any contaminated activity left onsite should be periodically monitored with the stipulation that it may be necessary to secure the contamination in a better fashion at a later date.

Contaminated soil that is transported offsite to a waste burial ground or the Federal Repository must be packaged in containers. This waste may be in a retrievable or non-retrievable form at the waste burial ground and in a retrievable form at the Federal Repository. The technological bases and the bases for estimating the costs of these techniques are discussed below.

(1) Fencing: The fences considered are 5 feet high, 6 feet high, and 6 feet high with three strands of barbed wire at the top of a galvanized or aluminum chain link fence. The costs range from about \$4 - \$8 per linear foot for each of the fences.[32-34] Assuming the land to be fenced is a perfect square, the average costs are about \$1,500/acre for a 10-acre plot, about \$500/acre for a 100-acre site, and about \$150/acre for a 1,000-acre plot. The ranges of costs are about \$400 - \$2,200 per acre for the 10-acre plot, \$300 - \$700 per acre for a 100-acre plot, and \$100 - \$225 per acre for a 1,000-acre plot. If a different geometry for the area is selected, the costs will range by a factor of 0.89 for a circle to 1.06 for a rectangle with the ratio of the sides of 2:1.

(2) Guard Service: A 24-hour guard service requires 5 guards for full time at approximately \$3.00 - \$5.00/hour.[45] At these assumed costs, the annual costs will range from about \$30,000 to about \$57,000 per year depending on the area of the country. For a specific case in Nevada, the annual costs for a contracted guard service is higher at \$100,000 for 3.8 man years per year.[53] Costs for guard service in areas that are not as remote as the case in Nevada will not be as high. If more than a single guard is required to be on duty (possibly due to the size of the area), then costs will be proportionately higher. Also, guard service cost at a specific site may be higher depending on higher local wage rates, overtime rates, specialized vehicles, and personnel that may be necessary, and other specialized restrictions.

(3) Waste Burial Grounds:[51,52] Solid radioactive waste with concentrations less than a Pu concentration of 10 nCi/gram are currently being buried at State burial grounds, such as the State-licensed ones in South Carolina, Nevada, Washington, Kentucky, New York, and Illinois. The one time charge for storing non-retrievable containerized wastes at these burial grounds range from about \$0.50 - \$2.00/cubic feet with an average of about \$1.35/cubic foot.[46-50] The one-time charges for retrievable wastes are estimated to be \$3 - \$5/cubic foot at these burial grounds, with an assumed value of \$4/cubic feet.[44]

(4) Federal Repository:[51,52] Higher level ($\geq$ 10nCi/gram) solid waste may be shipped to the Federal Repository, which is in the development stage. The outcome of this concept is dependent on rulemaking actions by the Nuclear Regulatory Commission. The Energy Research and Development Administration indicated the projected one-time charges to range from \$20 - \$30/cubic foot with an assumed value of about \$25/cubic foot.[47]

The one-time charges for burial at the waste burial grounds and acceptance at the Federal Repository include land costs, handling, repackaging, if necessary (for retrievable wastes), monitoring, inspection, and perpetual care. It is not known at this time if the charges indicated are adequate to defer all of the perpetual care costs of long-term burial of plutonium contaminated soils. The charges are either current charges or the best estimate of the costs.

The costs per cubic foot for storage is converted to costs per acre by assuming that 4 inches of soil are removed on each acre of land, or 538 cubic yards of soil are removed per acre.

Type of Waste Burial	Activity Level	$\$/\text{ft}^3$ Range	$\$/\text{ft}^3$ Average	$/Acre Assumed
Waste Burial Ground non-retrievable	<10nCi/gram	$ 0.50-$2	$ 1.35	$ 20,000
Waste Burial Ground retrievable	<10nCi/gram	$ 3-$5	$ 4.00	$ 60,000
Federal Repository retrievable	>10nCi/gram	$20-$30	$25.00	$360,000

Section G. Restoration

A necessary consideration in any discussion of the costs of a plutonium clean-up are the costs to restore the land after decontamination. These costs will vary, as do the costs discussed in other sections, with terrain, climate, soil type, the degree of contamination and most importantly with the desired end use of the land affected by the contamination. However, generally these costs would be comparable to the costs developed for recovery of coal strip-mined areas. Prior to restoration experts in this area should be consulted to determine the type and extent of restoration to be applied at a specific site.

Experience with recovery of plutonium-contaminated land has been limited primarily to "natural" restoration -- where man did not reestablish soil or vegetative cover but left the disturbed land to recover as best it could. More recently, the trend is toward restoring disturbed land to its original condition or to some productive status.

There are two main goals for any restoration effort on contaminated land: (1) to merely restore the vegetation or ground cover so that erosion does not occur or (2) to restore the surface so that both man and animals can have unrestricted access to it. Again, the factor determining which goal is selected will vary for every site, and it will be desirable from a cost standpoint if the goals are established prior to selection of a clean-up plan so that the decontamination can proceed with that goal in mind.

Obviously, the method of decontamination will determine the extent of the restoration procedures required. In those cases where the soil has been removed, it will be necessary to apply a new covering. The cost of a 4-inch soil covering, including delivery and distribution, will run between $1,500 - $3,000 an acre for loam, averaging $2,400 per acre, and $3,300 - $5,700 for topsoil, averaging $4,500 per acre.[4,28,29,31-34] These figures compare favorably with the cost of distributing an equal amount of soil at normalized costs at Enewetak -- $2,530 per acre.[30]

Following a logical sequence, the land could be treated to encourage revegetation. Lime and fertilizer could be added, at a cost of $20 - $480 per acre (averaging $150 per acre).[31,32-34] The land could then be seeded for $190 - $220 per acre (average $200 per acre).[31,32] In some cases, one of the chemical binders discussed in the section on Stabilization could be dispersed together with seed (at an additional cost of about $600 per acre) to hold them in place and protect the soil until a ground cover is established.[16]

Depending on the site, optional treatments might be selected. Mulch may be applied with the seed and binder (at a cost of from $35 - $190 per acre) to further protect the soil from erosion.[31,32,34,38] Grasses or legumes could be planted instead of the seeds for $20 - $220 per acre (averaging $100/acre).[31,38] Seedlings could be planted for $40 - $120 per acre (averaging $60 per acre).[38] Shrubs could also be substituted at a minimum installed cost of about $8.60 per shrub. Depending on the shrub density desired, costs would likely run from $860 per acre to $1,720 per acre.[34]

These costs and treatments apply under what might be called normal circumstances. A very special case for restoration would be desert conditions like those found at the Nevada Test Site. The sparseness of soil, vegetative cover, and water severely complicate the task of restoring a decontaminated area. Cost figures have not been computed for such an effort; but, research has been conducted to determine the most successful methods for restoring desert surfaces. Generally, the researchers concluded that due to the problems involved in such restoration, decontamination efforts which involve removal of soil or vegetation in a desert situation should be very carefully considered.[4]

As indicated in the section on Stabilizers, some stabilizing techniques include steps also considered necessary for restoration. There would be an obvious benefit from reduction of costs where the requirements for both can be met in a single operation.

APPENDIX A. Bases for Costs in Table 2

Part 2. Discussion of Specific Techniques*

I. Stabilization Only

1. Plowing

A. Radiological Surveillance - See Section A of Part 1.

B. Stabilization - First the area should be stabilized with a short-term stabilizer to reduce resuspension of plutonium to a minimum. The short-term stabilizer is the type used on ground that has vegetation, i.e., unplowed land or ground that is not bare soil. The costs of this type of stabilizer are discussed in Section B of Part 1. Then, the land can be plowed by farm machinery to a 12-inch depth or with a large plow pulled by bulldozers or heavy machinery to turn the top layers of soil under 3 feet of earth. This 3-foot depth should be sufficient to control uptake of plutonium by plants. Also, the plowing should help dilute the plutonium and remove plutonium from the surface of the land. The costs of a 3-foot plowing are similar to those for plutonium scraping in Appendix A, Part 2, Section 4A. A 12-inch plowing cost is in the range of a few tens of dollar an acre and is, thus, an insignificant cost.

C. Removal - Vegetation removal may be necessary prior to plowing. The types of removal that may be warranted are discussed in Section C of Part 1. There will be no soil removed from areas to be plowed.

D. Packaging - Not applicable. (See Section C of Part 1 concerning packaging of vegetation that has been removed from the land.)

E. Transportation - Not applicable.

F. Ultimate Disposal - Fencing in the plowed area may be necessary if the area is heavily populated and if the public should need to be excluded. The fence should also have the appropriate radiation warning signs. The need for fencing would be dependent on plutonium contamination and the effectiveness of the plowing operation.

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*Appendix A, Part 2, discusses costs for the specific techniques presented in Tables 1 and 2.

Temporary fencing may be necessary for the time period before the restoration procedures become effective. Guard service may also be necessary if exclusion of the public from the plowed area is essential. The costs of fencing and guard service are presented in Section F of Part 1.

G. Restoration - As a minimum, restoration for the plowed area would include fertilizing, seeding, and application of a short-term stabilizer. The stabilization costs presented in Table 2 are for the type of stabilizer that is used primarily for erosion control on bare ground as indicated in Table 6. The stabilizer should be adequate until vegetative growth becomes effective as a permanent stabilizer. The stabilizers are discussed in Section B of Part 1.

The alternative restoration procedures that may be necessary or desirable are mulching (instead of stabilization), starting a ground cover (instead of seeding), and placing seedlings or shrubs to aid in returning the land to its original state or to an acceptable status. Restoration costs are discussed in Section G of Part 1.

2. Chemical and/or Vegetative Stabilization

A. Radiological Surveillance - See Section A of Part 1.

B. Stabilization - As a preventive measure, a short-term stabilizer may be applied before any decision is made concerning the decontamination techniques to be employed. There are two types of stabilization discussed: (1) vegetation plus chemicals and (2) long-term (5 to 10 year life, as indicated in Table 6) chemical stabilization.

(1) Vegetation plus chemicals - A short-term stabilizer that is used primarily on bare ground as indicated in Table 6 is applied to the contaminated area to allow the vegetative ground cover to become effective. The short-term stabilizer costs are discussed in Section B of Part 1 and the vegetative ground cover costs are discussed in Section G of Part 1.

(2) Long-term stabilization - Chemical stabilizers that will be effective in stabilizing the soil for a 5 to 10 year period are applied. Reapplication may be necessary each 5 to 10 years or the soil may be decontaminated in the future using a different procedure. The long-term chemical stabilizer costs and types are discussed in Section B of Part 1.

C. Removal - Vegetation removal may be necessary prior to application of the stabilizers. The types of removal that may be warranted are discussed in Section C of Part 1. There will be no soil removed from the area to be stabilized by application of vegetation and/or chemicals.

D. Packaging - Not applicable. (See Section C of Part 1 concerning packaging of vegetation that has been removed from the land.)

E. Transportation - Not applicable.

F. Ultimate Disposal - Permanent fencing for the stabilized area would generally be more appropriate for this decontamination procedure than for any of the other procedures or techniques. A discussion of fencing and guard service is presented in Section 1.F of Part 2 and Section F of Part 1.

G. Restoration - Not applicable.

3A. Soil Cover as a Stabilizer - 4 inches of soil.

A. Radiological Surveillance - See Section A of Part 1.

B. Stabilization - The area to be covered with soil should be stabilized with the short-term stabilizer as indicated in Table 6 that is used on land with vegetation. This reduces resuspension prior to and during application of the soil. There are two choices of soil - loam and topsoil, with loam usually being adequate for most decontamination purposes. The costs of soil application are directly proportional to the soil depth. Some types of land may not be appropriate for placing a soil cover depending on soil, type, terrain, moisture in the soil, etc. The costs of the soil, its application, spreading, etc., are presented in Section B of Part 1. If this type of stabilization is not effective, other methods may have to be utilized at a future date.

C. Removal - Vegetation removal may be necessary prior to application of the soil. The types of removal that may be warranted are discussed in Section C of Paft 1. There will be no soil removed from areas to be stabilized by application of soil.

D. Packaging - Not applicable. (See Section C of Part 1 concerning packaging of vegetation that has been removed from the land.)

E. Transportation - Not applicable.

F. Ultimate Disposal - A discussion of fencing and guard service is presented in Section 1.F of Part 2 and Section F of Part 1. Permanent fencing and guard service is probably not appropriate but temporary fencing may be used until restoration procedures become effective.

G. Restoration - The restoration procedures appropriate for a 4-inch soil cover are identical to those discussed in Section 1.G of Part 2. Restoration costs are discussed in Section G of Part 1.

3B. Soil Cover as a Stabilizer - 12 inches of soil.

A. Radiological Surveillance - See Section A of Part 1.

B. Stabilization - The procedures are the same as those presented in Section 3A.B. in Part 2 except 12 inches of loam are spread or 8 inches of loam with 4 inches of topsoil are spread. The extra depth of soil (8 inches extra) would be for added stabilization for reducing resuspension of soil, and to reduce uptake of plutonium by plants, thus allowing the land to be used productively. (The choice of 12 inches of soil is primarily for calculational purposes.) The costs of applying the soil are directly proportional to the depth of the soil and are discussed in Section B of Part 1. If this type of decontamination is not effective, other methods may have to be used in the future.

C. Removal - Vegetation removal may be necessary prior to application of the soil. The types of removal that may be warranted are discussed in Section C of Part 1. There will be no soil removed from areas to be stabilized by application of soil.

D. Packaging - Not applicable. (See Section C of Part 1 concerning packaging of vegetation that has been removed from the land.)

E. Transportation - Not applicable.

F. Ultimate Disposal - The soil cover, after restoration procedures become effective, should be sufficient in itself as an ultimate disposal. Fencing and a guard service should not be necessary since the soil covering would be about a foot deep; thus, it is unlikely erosion could remove the entire soil covering.

G. Restoration - The procedures for restoring the land should be identical to those discussed in Section 1.G of Part 2.

II. Removal with Onsite Disposal

4A. Scraping into Windrows.

A. Radiological Surveillance - See Section A of Part 1.

B. Stabilization - Prior to soil removal, the land should be stabilized with a short-term stabilizer (as indicated in Table 6) to reduce resuspension of plutonium to a minimum. The cost of this type of stabilizer is discussed in Section B of Part 1.

The windrows are stabilized in the same manner as the land in Section 2.B of Part 2 using long-term chemical stabilizers or a combination of chemical and vegetative stabilizers. The long-term chemical stabilizers may need to be reapplied every 5 to 10 years. If this technique is evaluated at a future date to be unacceptable, the windrows may be scraped up and buried onsite or transported offsite for burial. The costs of the stabilizers are discussed in Section B of Part 1.

C. Removal - Vegetation removal will probably be necessary prior to scraping the land into windrows. The types of removal that may be warranted are discussed in Section C of Part 1.

Soil removal will be by a scraper, grader or dozer with an angled blade to produce windrows that are assumed to be about 18 inches high and 3-1/2 feet wide at the base.[19] The depth of soil removed by scraping is assumed to be 4 inches. The costs will vary considerably with the type of soil and the terrain. Soil removal is also discussed in a little more detail in Section C of Part 1. The range of costs range from less than $100 per acre for sandy soils with ideal conditions to six or seven hundred dollars per acre for clayey hard soils during adverse conditions. The average costs from the data available are calculated to be about 200 dollars per acre except for sand (about 70 dollars per acre).[3-4,16,33-35,37-40]

D. Packaging - Not applicable. (See Section C of Part 1 concerning packaging of vegetation that has been removed from the land.)

E. Transportation - Transportation costs would be incurred only if the windrows are pushed several thousand feet. The extra costs for handling or pushing soil with a scraper are discussed in Section E of Part 1.

F. Ultimate Disposal - A discussion of fencing and guard service is presented in Section 1.F of Part 2 and Section F of Part 1. Permanent fencing and guard service would probably not be necessary, especially after restoration procedures become effective.

G. Restoration - The windrows are stabilized with chemicals and/or vegetation and are not usually restored further. The windrows will cover approximately 50 percent of the land area. Thus, the remaining land area would have to be restored by applying (1) a soil cover of loam (assumed to be 4 inches) to replace the soil that was scraped, (2) short-term stabilizer to cover bare ground (as indicated in Table 6), (3) fertilizer, (4) seeds, and probably (5) lime. The restoration costs are based on restoring 50 percent of each acre since 50 percent of an acre is covered with windrows and will not be restored. The stabilizer is discussed in Section B of Part 1, and the restoration procedures are discussed in Section G of Part 1.

4B. Scraping into Mounds

A. Radiological Surveillance - See Section A of Part 1.

B. Stabilization - Stabilization techniques for scraping the soil into mounds are the same as those discussed for scraping the soil into windrows in Section 4A.B of Part 2.

C. Removal - Vegetation removal will probably be necessary prior to scraping the land to form mounds. The types of removal that may be warranted are discussed in Section C of Part 1.

Soil removal would be by a scraper, grader, or dozer that can either push or carry the soil a short distance to make mounds. It is assumed that a nominal haul distance is 1,000 feet and the mound size is 10 feet high with a base of 50 feet by 100 feet. The mound is assumed to have sides that slope at a 45 degree angle and a flat surface at the top that is 80 feet long and 30 feet wide. (The shape and dimensions of the mound were chosen for calculational purposes to determine the surface area a mound would cover.) If 4 inches of soil are removed from the land, the mounds would require only about 5 percent of the surface area per acre.

The range in costs for scraping and hauling the soil about 1,000 feet to form a mound were calculated to be about $150 to $1,640 per acre. The average cost is about $600 per acre.[3-4,16,33-35,37-40]

D. Packaging - Not applicable. (See Section C of Part 1 concerning packaging of vegetation that has been removed from the land.)

E. Transportation - The transportation costs indicated would be incurred if the distance the soil must be hauled to the mounds is greater than 1,000 feet. The extra costs for hauling (or possibly pushing) the soil with a scraper are discussed in Section E of Part 1.

F. Ultimate Disposal - The procedures would be similar to those used in Section 4A.F in Part 2.

G. Restoration - The restoration costs only apply to the area of land from which the soil is removed. This area is calculated to be 95% of the original area since the mounds cover about 5% of the land area. This calculation is based on the assumption of the geometry of the mounds as presented in Appendix A, Part 2, Section 4B.C. The techniques and procedures would be identical to those in Section 4A.G in Part 2 except the costs would be calculated for 95% of an acre instead of the 50% of an acre for scraping land into windrows. More details on costs are presented in Section G of Part 1.

4C. Scraping and Hauling Soil into Trenches (includes a 1,000 foot transport distance)

A. Radiological Surveillance - See Section A of Part 1.

B. Stabilization - Prior to soil removal, the land should be stabilized with a short-term stabilizer (as indicated in Table 6) to reduce resuspension of plutonium to a minimum. The cost of this type of stabilizer is discussed in Section B of Part 1.

C. Removal - Vegetation removal will probably be necessary prior to scraping the land. The types of removal that may be warranted are discussed in Section C of Part 1.

Soil removal would be by a scraper, grader, or dozer that can carry (or possibly push) the soil a short distance to the trenches. The trenches are dug and backfilled by large earth moving machinery. The soil removed from the trenches may be applied to the scraped land to help defer the soil costs for restoration of the scraped land. The costs of burying the contaminated soil include a 4-inch removal of soil and a 1,000-foot haul (or possibly push) of the soil to the trench. The range of costs is estimated to be $530 - $3.710 per acre with an average of about $1,700 per acre.[3-4,16,33-35,37-40] If the trenches are constructed near the scraped areas, about $300 per acre in hauling (transportation) costs can be eliminated.[32,33,34]

D. Packaging - Not applicable. (See Section C of Part 1 concerning packaging of vegetation that has been removed from the land.)

E. Transportation - The transportation costs indicated would be incurred if the distance the soil must be hauled to the trenches is greater than 1,000 feet. The extra costs for hauling or pushing the soil with a scraper are discussed in Section E of Part 1.

F. Ultimate Disposal - Fencing and a guard service around the scraped land or the trenches is probably not necessary since the contaminated soil is buried.

G. Restoration - Restoration procedures would be the same as those discussed in Section 1.G of Part 2 with one exception: the soil from the trenches can be used to cover the scraped area; thus, there is no cost of procuring the soil. An option may be to add 2 inches of topsoil to promote vegetation growth.

5. Vacuuming with Disposal of the Soil in a Trench

The techniques and costs are similar for Vacuuming (with trench disposal) and Scraping (with trench disposal) for all the categories except for (1) stabilization and (2) soil removal.

(1) Stabilization - There is little available data concerning stabilization prior to vacuuming. Temporary stabilizers used on bare ground (as indicated in Table 6) are assumed to be adequate prior to vacuuming. However, the use of stabilizers prior to vacuuming may interfere with this process. The land area will probably require plowing or discing to break up the stabilizer. Even water used as a stabilizer might form a crust which can impede vacuuming.

(2) Soil Removal - There is limited information concerning the use of an industrial vacuum cleaner for soil removal. Sand can be picked up with a high efficiency and with ease.[1] Most other types of soil should be shallow plowed (several inches depth) prior to vacuuming to loosen the deep soil. Plowing (8-10 inches depth or deeper) will move the contamination below grade thus reducing the efficiency of vacuuming. The efficiency for removal for these soils is less than sand and is very dependent on the moisture content of the soil.

The vacuum cleaner must have a HEPA filter for preventing plutonium resuspension in the air. The vacuum cleaner would be mounted on a boom on a flatbed truck. The soil removal costs of this technique a 4-inch removal were calculated for plowing, vacuuming, building a trench, and covering the trench. The range of costs were estimated to be $910 - $5,100 per acre with the average assumed to be about $2,500 per acre.[1]

III. Removal with Offsite Disposal

6A & 6B. Scraping or Vacuuming

A. Radiological Surveillance - See Section A of Part 1.

B. Stabilization - For scraping, stabilization is the same as that discussed in Section 4.C.B. For vacuuming, stabilization is the same as that discussed in Section 5.

C. Removal

(1) The soil is plowed, vacuumed, and deposited in containers to be transported offsite. This is discussed further in Section C of Part 1.

(2) An alternative method with a similar cost would be to scrape the soil into windrows or mounds, pick up the soil using an industrial vacuum cleaner, and deposit the soil in a container to be transported offsite. It may be easier and more economical to pick up the soil with a scraper or a front end loader to deposit the soil in containers. With either method, it is very important to keep spillage of soil to a minimum.

If the soil is picked up manually, rather than by a vacuum cleaner, the costs for the pickup alone can range between $2,000 - $6,000 per acre[33,34] for a 4-inch removal. Thus vacuuming seems to be economical, as well as fast.

D. Packaging - The packaging costs are discussed in Section D of Part 1 for the Waste Burial Grounds and for Federal Repositories.

E. Transportation - The transportation of containerized soil is discussed in Section E of Part 1.

F. Ultimate Disposal - Ultimate disposal is discussed for the Waste Burial Grounds and the Federal Repository in Section F of Part 1.

G. Restoration - The restoration of the land includes the cost of materials (including 4 inches of soil) and their application for a full acre. A discussion of the costs for restoration are presented in Section G of Part 1.

APPENDIX B. Special Techniques

Part 1 - Technology

There are several techniques not listed in Table 1 that may be used for reducing the environmental impact of plutonium that has been deposited on land areas. These techniques are generally used for the reasons indicated below.

(1) Application of Asphalt: A surface treatment of asphalt can be applied on small areas as a permanent stabilizer.[5] This type of surface would not be adequate as a parking area, but it would effectively prevent resuspension of plutonium (100% efficient) and for a number of years prevent vegetation growth (about 100% efficient). This method probably would not be used because of the high costs and the commitment of the land to a nonproductive status. The stabilizer action and its costs are discussed in more detail in Appendix A, Part 1, Section B.

(2) Sewage Sludge: Sludge may be applied to land as a fixation stabilizer just as soil is applied (as described in Appendix A, Sections 3A and 3B of Part 2) or in lieu of restorative measures requiring soil and vegetative applications. The savings in cost would be the material cost of soil. However, this method would only be applicable to areas where large quantities of sludge are available and the land can be committed as a sewage sludge disposal site.

(3) Snow: Decontamination of snow probably would involve collecting and melting the snow, and then removing the plutonium by evaporation or ion exchange, and disposal at a waste burial ground. The solid waste would be about a factor of 100 smaller* than the volume of snow scraped, which would significantly reduce the costs of packaging and burying the waste. Thus, scraping the snow with ultimate disposal at an approved waste burial site could be less expensive than scraping and burying a similar volume of soil, even though there would be additional costs for solidification or ion exchange.

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*Melting snow to water reduces the volume by about a factor of 8 to 12.[12] Present day techniques of solidification can reduce a volume of contaminated water by a factor of 10 or more.[45] Solidification of the waste would then probably increase this volume by a factor of 2 or more.

(4) Flooding land areas with about an inch of water, combined with collection of the water, removes loose soil and debris containing Pu. This technique would result in large volumes of contaminated water requiring disposal. Since many waste burial grounds and the Federal Repository probably will not bury or store liquids, the contaminated liquids would also require solidification. Flooding might be useful, even though difficult, on mountainous areas which are inaccessible to large earthmoving equipment. However, for most land areas, flooding would not be acceptable because of the generation of large volumes of contaminated water and the difficulties of collection of the contaminated water.

(5) Air Shelters: A canvas type structure supported by air is presently being utilized at the Idaho National Engineering Laboratory to allow the excavation of solid waste burial drums during advserse weather conditions. A similar type structure covering approximately half an acre can be utilized during plutonium decontamination operations to help prevent the wide disposal of plutonium contaminated dust particles (i.e. resuspension) and/or to provide protection from the weather. These shelters are large enough to allow the limited movement and operation of scrapers, dozers, and other earthmoving equipment within the structure. The structure can also be transported from one area to the other so a reasonably large area (several acres to several tens of acres) may be decontaminated.[55,56]

(6) Other measures: Other measures to reduce the environmental impact of plutonium deposited on farmland could possibly be (1) the use of additives, such as lime or fertilizer, to reduce the entry of radioactivity from soils into crops; (Note: Additives to reduce plant uptake of plutonium were not specifically mentioned in the literature. It should also be noted that there is no information in the literature to indicate that plutonium in the forms which might normally be distributed is taken up in growing crops to a significant degree.) (2) growing crops that take up small amounts of plutonium; or (3) treating of milk or other products during the processing stages for removal of plutonium;[6] (4) leaching out plutonium contamination from the soil by some chemical treatment method, and (5) selectively screening contaminated soil so as to separate fine particulates from stones and larger material with the idea that most of the plutonium distribution will reside with the small particle fraction.

Part 2 - Costs

(1) Application of Asphalt: See Appendix A Section B of Part 1 concerning costs of a surface treatment of asphalt.

(2) Sewage Sludge: Costs of hauling and spreading sewage treatment plant sludge are calculated to range between about \$200 -\$3,800/acre/year. The sludge could be spread for several years. The bases of these calculations are presented on page 9 and Tables 9 and 10 of reference 24.

(3) Snow: The costs for scraping, packaging, and disposing of contaminated snow are assumed to be proportional to those for soil, except the volume of solid waste per acre is assumed to be a factor of about 100 smaller. These assumptions are made to present a ball-park estimate and other costs such as demineralization or evaporation of the melted snow are not included. The range of costs is estimated by dividing by 100 the costs presented in 6A and 6B of the "typical" project discussed in Appendix B. These costs are \$900 - \$5,100/acre.

(4) Flooding (1 inch of water): The costs to flood an acre of land with 1 inch of water would be proportional to the costs for applying water as a stabilizer (See Appendix A, Section B, Part 1). The costs of flooding would include radiological surveillance, packaging (at \$6.80 - \$10.20 per ft^3 or an extra \$49,000 - \$74,000/acre, See Appendix A, Section D, Part 1), Ultimate Disposal and Restoration. The costs for applying the water will be about (1.0/0.3) \$220/acre = \$730/acre and a range of about \$330 - \$1,130/acre.[16,42] The water that is collected would probably have to be solidified by the same techniques as used for melted snow, with disposal of the solid material at a waste burial ground or the Federal Repository. As an example of the disposal cost, assume that the water is encased in concrete with an increase in volume of a factor of 2. The volume of concrete would be 7260 ft^3. At a waste burial ground, the disposal costs would be about \$9800/acre for nonretrievable disposal, \$29,000/acre for retrievable storage, and about \$182,000/acre for storage at the Federal Repository. (See Appendix A, Section F Part 1). If the volume of solid material is significantly less (as a result of using another solidification technique), then the packaging and disposal costs can also be reduced significantly.

(5) Air Shelters: The one time direct and indirect costs for an air shelter, including anchor blocks is estimated to be about \$170,000 (1975 dollars). This shelter can enclose about 22,500 square feet. The costs to initially install the structure or to move and install the structure near its original installation is about \$49,000 (1975 dollars).[56]

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TABLE 1 PLUTONIUM DECONTAMINATION

<table>
<tr><td>DECONTAMINATION TECHNIQUES →</td><td colspan="4">I STABILIZATION ONLY</td><td colspan="4">II REMOVAL WITH ONSITE DISPOSAL</td><td colspan="2">III REMOVAL WITH OFFSITE DISPOSAL AT A LICENSED BURIAL GROUND</td></tr>
<tr><td>STEPS INVOLVED IN CLEANUP EFFORT ↓</td><td>1 PLOWING</td><td>2 CHEMICAL VEGETATIVE</td><td>3A SOIL COVER - 4"</td><td>3B SOIL COVER - 12"</td><td>4A SCRAPING INTO WINDROWS</td><td>4B SCRAPING INTO MOUNDS</td><td>4C SCRAPING INTO TRENCHES</td><td>5 VACUUMING WITH TRENCH DISPOSAL</td><td>6A SCRAPING</td><td>6B VACUUMING</td></tr>
<tr><td>A. RADIOLOGICAL SURVEILLANCE</td><td colspan="10">SURVEILLANCE REQUIRED BEFORE, DURING AND AFTER DECON PROJECT. RECOMMENDATIONS OFFERED IN SECTION A, APPENDIX A</td></tr>
<tr><td rowspan="2">B. STABILIZATION</td><td rowspan="2">(SHORT TERM STABILIZER APPLIED PRIOR TO PLOWING FOR DUST CONTROL)
SHALLOW PLOWING (1) - REGULAR FARM EQUIPMENT TO PREVENT RESUSPENSION - OR -
DEEP PLOWING - (2) - HEAVY EQUIPMENT TO TURN SURFACE ACTIVITY AND TO DILUTE ACTIVITY IN SOIL</td><td rowspan="2">CHEMICAL AND VEGETATIVE
SHORT TERM CHEMICAL STABILIZER APPLIED FOR DUST CONTROL AND TO HELP PREVENT EROSION. VEGETATION - SEEDLINGS, GROUND COVER, SHRUBS - OR -
LONG TERM CHEMICALS LASTING 5-10 YRS WITH REAPPLICATION OR SCRAPING AT LATER DATE</td><td colspan="2" rowspan="2">(SHORT TERM STABILIZER APPLIED PRIOR TO SOIL SPREADING FOR DUST CONTROL.)
SOIL COVER SERVES AS MORE PERMANENT STABILIZER</td><td colspan="6">SHORT TERM STABILIZER APPLIED PRIOR TO REMOVAL ACTIVITIES TO CONTROL DUST AND HELP PREVENT RESUSPENSION</td></tr>
<tr><td colspan="2">LONG TERM STABILIZER &/OR VEGETATION TO FIX THE WINDROWS / THE MOUNDS</td><td colspan="4">STABILIZERS AS APPROPRIATE TO ASSIST IN RESTORATION PROCESS. SEE "RESTORATION" BELOW</td></tr>
<tr><td rowspan="3">C. REMOVAL</td><td colspan="10">VEGETATION REMOVAL WHERE APPROPRIATE</td></tr>
<tr><td colspan="4" rowspan="2">SOIL REMOVAL NOT ANTICIPATED</td><td colspan="6">PLANNED SOIL REMOVAL</td></tr>
<tr><td>METHOD: SCRAPERS AND DOZERS</td><td>METHOD: SCRAPERS AND DOZERS</td><td>METHOD: SCRAPERS AND DOZERS - SOIL THEN TRUCKED TO TRENCHES</td><td>METHOD: PLOW THEN VACUUM THEN TRUCK TO TRENCHES</td><td>METHOD: SCRAPE AND PICK UP</td><td>METHOD: PLOW AND VACUUM</td></tr>
<tr><td>D. PACKAGING</td><td colspan="8">NOT APPLICABLE</td><td colspan="2">IF SENT TO A ① STATE BURIAL GROUND - MATERIAL WILL BE PACKAGED IN LINED, 55-GALLON DRUMS. (WILL INCLUDE LOWER ACTIVITY CONTAMINANT.)
"OR" A ② FEDERAL REPOSITORY - MATERIAL WILL BE PACKAGED IN LINED, 55-GALLON DRUMS. (HIGHER LEVEL OF ACTIVITY AND HIGHER ASSOCIATED COSTS.)
SPECIAL CASE: POLYURETHANE FOAM CAN BE CRUSHED AND PACKAGED IN DRUMS OR INCINERATED AND DRUMMED.</td></tr>
<tr><td>E. TRANSPORTATION</td><td colspan="5">NOT APPLICABLE</td><td>TRANSPORTATION COSTS VARY WITH PUSH DISTANCE BETWEEN SCRAPED AREA AND MOUND (COSTS FOR THE FIRST THOUSAND FEET ARE INCLUDED IN THE REMOVAL COSTS.)</td><td colspan="2">SAME AS 4-B EXCEPT SOIL MAY BE LOADED AND TRANSPORTED TO TRENCH. (AGAIN COSTS FOR THE FIRST THOUSAND FEET ARE INCLUDED IN THE REMOVAL COSTS.)</td><td colspan="2">INCLUDES ONLY TRANSPORTATION COSTS FOR DISTANCES IN EXCESS OF ABOUT [illegible] MILES. (COSTS FOR TRANSPORTATION OF THE CONTAINMENTS UP TO THESE DISTANCES ARE INCLUDED IN THE PACKAGING COSTS DISCUSSED ABOVE.)</td></tr>
<tr><td rowspan="3">F. ULTIMATE DISPOSAL</td><td colspan="8">DISPOSAL - STABILIZED ONSITE</td><td colspan="2" rowspan="3">DISPOSAL SITE DEPENDS ON ACTIVITY OF MATERIAL. LOW LEVEL WASTES CAN BE STORED IN NONRETRIEVABLE FORM - HIGH LEVEL ARE STORED IN RETRIEVABLE FORM. ACCORDINGLY, THREE OPTIONS ARE AVAILABLE:
1 STATE BURIAL GROUND - NON-RETRIEVABLE
2 STATE BURIAL GROUND - RETRIEVABLE
3 FEDERAL REPOSITORY - RETRIEVABLE
} ALL INVOLVE PERPETUAL CARE</td></tr>
<tr><td>OPTIONAL
FENCING AND GUARD SERVICE</td><td>REQUIRED (PROBABLY)
FENCING AND GUARD SERVICE</td><td>OPTIONAL
FENCING AND GUARD SERVICE</td><td>PROBABLY NOT NECESSARY
FENCING AND GUARD SERVICE</td><td colspan="4">(IN WINDROWS) (IN MOUNDS)
OPTIONAL
FENCING AND GUARD SERVICE</td></tr>
<tr><td colspan="8">FURTHER DECONTAMINATION MAY BE NECESSARY</td></tr>
<tr><td>G. RESTORATION</td><td>RESTORATION AS NECESSARY
ADDITIONAL SOIL NOT NEEDED
STABILIZER APPLIED FOR EROSION CONTROL</td><td>NOT APPLICABLE</td><td colspan="2">RESTORATION NECESSARY TO HOLD SOIL COVER IN PLACE</td><td>RESTORATION NECESSARY FOR SCRAPED AREAS NOT COVERED BY WINDROWS. BEGINS WITH ADDITION OF SOIL COVER. WINDROWS MAINTAINED AS INDICATED ABOVE</td><td>RESTORATION NECESSARY FOR SCRAPED AREAS NOT COVERED BY MOUNDS - BEGINS WITH ADDITION OF SOIL COVER. MOUNDS MAINTAINED AS INDICATED ABOVE</td><td>SOIL REMOVED FROM TRENCHES CAN BE DISTRIBUTED OVER SCRAPED AREAS - LITTLE ADDITIONAL SOIL SHOULD BE NECESSARY. OTHER RESTORATION AS INDICATED IN SECTION G, APPENDIX A</td><td>SOIL REMOVED FROM TRENCHES CAN BE DISTRIBUTED OVER VACUUMED AREAS - LITTLE ADDITIONAL SOIL SHOULD BE NECESSARY. OTHER RESTORATION AS INDICATED IN SECTION G, APPENDIX A</td><td colspan="2">COMPLETE RESTORATION REQUIRED. PROCEED AS INDICATED IN SECTION G, APPENDIX A.</td></tr>
</table>

TABLE 2 DECONTAMINATION COSTS

DECONTAMINATION TECHNIQUES	STABILIZATION ONLY				REMOVAL WITH ONSITE DISPOSAL				REMOVAL WITH OFFSITE DISPOSAL AT A LICENSED BURIAL GROUND	
STEPS INVOLVED IN CLEANUP EFFORT	1 PLOWING	2 CHEMICAL VEGETATIVE	3A SOIL COVER 6"	3B SOIL COVER 12"	4A SCRAPING INTO WINDROWS	4B SCRAPING INTO MOUNDS	4C SCRAPING INTO TRENCHES	5 VACUUMING WITH TRENCH DISPOSAL	6A SCRAPING	6B VACUUMING
A. RADIOLOGICAL SURVEILLANCE	(RANGE $750 $1100/A) (ASSUMED VALUE $900/A) (COSTS ARE ESTIMATED FOR A 3 MONTH PROJECT)									
B. STABILIZATION	SHORT TERM STABILIZER PRIOR TO PLOWING (RANGE $200 $1200/A) (ASSUME $700/A) DEEP PLOWING (RANGE $20 $[illegible]) (ASSUME $[illegible]) SHALLOW PLOWING RANGE $20 $85 ASSUME $45	SHORT TERM STABILIZER FOR DUST CONTROL (RANGE $200 $1200/A) (ASSUME $700/A) VEGETATION RANGE $20 270/A ASSUME $100 ($200 $1470) OR $800/A) OR LONG TERM CHEMICAL RANGE $520 $[illegible] ASSUME $[illegible]/A	SHORT TERM STABILIZER AND LOAM (RANGE $1700 $4500/A) (ASSUME $3100/A) SHORT TERM STABILIZER AND TOPSOIL RANGE $2900 $[illegible]/A ASSUME $6200/A	SHORT TERM STABILIZER AND 12" LOAM (RANGE $4700 $11 100/A) (ASSUME $7 900/A) SHORT TERM STABILIZER AND 8" LOAM & TOPSOIL RANGE $[illegible] $17 500/A ASSUME $10 000/A	A) SHORT TERM STABILIZER APPLIED PRIOR TO SCRAPING OR VACUUMING FOR DUST CONTROL (RANGE $200 $1200/A) (ASSUME $700/A) B) LONG TERM STABILIZER ON WINDROWS RANGE $[illegible] $330 ASSUME $200/A OR B) VEGETATION (RANGE $110 $710) (ASSUME $400/A)	B) LONG TERM STABILIZER ON MOUNDS RANGE $25 $35/A ASSUME $30/A OR B) VEGETATION (RANGE $10 $70) (ASSUME $20/A)	B) SEE RESTORATION			
C. REMOVAL	A) VEGETATION REMOVAL (OPTIONAL) GRASS COVER – RANGE $35 $750 ASSUME $[illegible]/A SLIGHTLY WOODED – RANGE $820 $1[illegible] ASSUME $1750/A HEAVILY WOODED – RANGE $750 $3400 ASSUME $2000/A NO SOIL REMOVAL COSTS				B) SOIL REMOVAL (RANGE $30 $[illegible]/A) (ASSUME $200/A)	B) SOIL REMOVAL (RANGE $150 $1640) (ASSUME $[illegible]/A)	B) SOIL REMOVAL (RANGE $530 $3700/A) (ASSUME $1700/A)	B) SOIL REMOVAL (RANGE $810 $5100) (ASSUME $2500)	B) SOIL REMOVAL (RANGE $450 $2800/A) (ASSUME ~$800/A)	B) SOIL REMOVAL (RANGE $[illegible] $[illegible]) (ASSUME ~$1000/A)
D. PACKAGING	NO PACKAGING COSTS INVOLVED								IF MATERIAL IS TO BE SENT TO A OR A 1) STATE BURIAL GROUND – (RANGE $[illegible] $[illegible]/A) (ASSUME ~$[illegible]/A) 2) FEDERAL REPOSITORY – (RANGE $[illegible] $[illegible]/A) (ASSUME ~$[illegible]/A) (NOTE: COSTS INCLUDE DRUMMING, LOADING, TRUCK FREIGHT CHARGES AND UNLOADING AT BURIAL GROUND)	
E. TRANSPORTATION	NO TRANSPORTATION COSTS INVOLVED				COSTS FOR FIRST 100 FT OF ONSITE HAUL INCLUDED IN REMOVAL COSTS. EACH ADDITIONAL 1000 FT OF HAUL – RANGE $[illegible] $[illegible]/A ASSUME $[illegible]/A				FOR MILEAGE IN EXCESS OF 1000 TO 1200 MILES COSTS ARE ADDED PER ADDITIONAL 100 MILES FOR EACH ADDITIONAL 100 MILES RANGE $2700 $10 000/A ASSUME $[illegible]/A	
ULTIMATE DISPOSAL	FENCING AND GUARD SERVICE MAY OR MAY NOT BE NEEDED DEPENDING ON CIRCUMSTANCES. IF NEEDED: FENCING FOR 10 ACRES – $1500/A RANGE $800 2200 100 ACRES – $500/A RANGE $300 700 GUARD SERVICE – $30 000 $57 000/YEAR								IF THE MATERIAL IS TO BE MAINTAINED IN: 1) NON-RETRIEVABLE FORM AT A STATE BURIAL GROUND (RANGE $7 000 $20 000/A) (ASSUME ~$[illegible]/A) OR 2) RETRIEVABLE FORM AT A STATE BURIAL GROUND (RANGE $44 000 $73 000/A) (ASSUME $[illegible]/A) OR 3) RETRIEVABLE FORM AT A FEDERAL REPOSITORY (RANGE $20[illegible]/A) (ASSUME $[illegible]/A)	
G. RESTORATION	RANGE / ASSUME (NO SOIL COSTS) FERT & LIME $20 400 $150/A STABILIZER $200 1200 $720/A SEEDING $190 220 $200/A $410 1820 $1060/A OPTIONS OR ALTERNATIVES MULCH $35 180 $125/A GROUND COVER $10 220 $100/A SEEDLINGS $40 120 $80/A SHRUBS $[illegible] 1720 $1300/A	RESTORATION COSTS INCLUDED UNDER STABILIZATION	RANGE / ASSUME (NO SOIL COSTS) FERTILIZER $20 400 $150/A TO STABILIZE $200 1200 $570/A SEEDING $190 220 $200/A $410 1880 $1060/A OPTIONS OR ALTERNATIVES LIME $20 $20/A MULCH $35 180 $125/A GROUND COVER $20 220 $100/A SEEDLINGS $40 120 $80/A SHRUBS $880 1720 $1300/A	RANGE / ASSUME (NO SOIL COSTS) FERTILIZER $20 400 $150/A STABILIZER $200 1200 $570/A SEEDING $190 220 $200/A $410 1880 $1060/A OPTIONS OR ALTERNATIVES LIME $20 $20/A MULCH $35 180 $125/A GROUND COVER $20 220 $100/A SEEDLINGS $880 1720 $1300/A SHRUBS $40 120 $80/A	(RESTORATION COSTS ASSUMING ~50% OF AN ACRE WILL REQUIRE SOIL) RANGE / ASSUME LOAM $750 1850 $1200/A FERTILIZER $10 240 $75/A LIME $20 $20/A STABILIZER $100 600 $285/A SEEDING $95 110 $100/A ($975 2820) ($1680/A) OPTIONS OR ALTERNATIVES TOPSOIL $1660 2850 $2250/A MULCH $20 95 $65/A GROUND COVER $10 110 $50/A SEEDLINGS $20 60 $30/A SHRUBS $430 860 $660/A	(RESTORATION COSTS ASSUMING ~90% OF AN ACRE WILL REQUIRE SOIL) RANGE / ASSUME LOAM $1425 3135 $2280/A FERTILIZER $20 400 $190/A LIME $20 $20/A STABILIZER $180 1140 $560/A SEEDING $180 210 $190/A ($1835 4885) ($3190/A) OPTIONS OR ALTERNATIVES TOPSOIL $3135 5415 $4275/A MULCH $30 180 $120/A GROUND COVER $20 210 $95/A SEEDLINGS $35 110 $80/A SHRUBS $820 1635 $1225/A	(ASSUMING SOIL FROM TRENCHES IS USED SO ONLY COST IS FOR APPLICATION) RANGE / ASSUME SOIL APPLICATION $700 880 $830/A FERTILIZER $20 400 $190/A LIME $20 $20/A STABILIZER $200 1200 $570/A SEEDING $180 210 $190/A ($1210 2770) ($1780/A) OPTIONS OR ALTERNATIVES TOPSOIL $1660 2850 $2250/A MULCH $35 180 $125/A GROUND COVER $20 220 $100/A SEEDLINGS $40 120 $80/A SHRUBS $880 1720 $1300/A	(ASSUMING SOIL FROM TRENCHES IS USED SO ONLY COST IS FOR APPLICATION) RANGE / ASSUME SOIL APPLICATION $700 880 $830/A FERTILIZER $20 400 $190/A LIME $20 $20/A STABILIZER $200 1200 $570/A SEEDING $180 210 $190/A ($1210 2770) ($1780/A) OPTIONS OR ALTERNATIVES TOPSOIL $1660 2850 $2250/A MULCH $35 180 $125/A GROUND COVER $20 220 $100/A SEEDLINGS $40 120 $80/A SHRUBS $880 1720 $1300/A	RANGE / ASSUME LOAM $1900 3300 ~$2600/A FERTILIZER $20 400 ~$190/A LIME $20 ~$20/A STABILIZER $200 1200 ~$570/A SEEDING $180 210 ~$190/A OPTIONS OR ALTERNATIVES ($[illegible] [illegible]) (~$[illegible]/A) TOPSOIL $3300 5700 ~$4500/A MULCH $35 180 ~$125/A GROUND COVER $20 220 ~$100/A SEEDLINGS $40 120 ~$80/A SHRUBS $880 1720 ~$1300/A	
TOTAL PROJECT COSTS ASSUMED AVERAGES RANGES (NOTE: VEGETATION REMOVAL COSTS AND FENCING/GUARD SERVICE COSTS NOT INCLUDED) (COSTS IN BRACKETS ARE THOSE USED TO CALCULATE TOTALS)	$2800/A $[illegible]/A	$2100/A $500 [illegible]	$4800/A $2400 7500/A	$[illegible] $5400 14 000	$3800 $1600 6300	$5100 $2400 8800	$[illegible] $1200 [illegible]	$[illegible] $3890 10 200	STATE BURIAL GROUND NON RETRIEVABLE AV $125 000 R $110 000 $[illegible]/A STATE BURIAL GROUND RETRIEVABLE AV $165 000 R $150 000 $230 000/A FEDERAL REPOSITORY RETRIEVABLE AV $515 000 R $[illegible] $[illegible]/A	

TABLE 3

Adjustment Indexes*

Multiply the costs appearing in Table 2 by the following factors to adjust for geographical differences:

Location	Labor	Material
Albany, New York	.96	1.01
Albuquerque, New Mexico	.84	.93
Anchorage, Alaska	1.29	1.04
Atlanta, Georgia	.82	.89
Austin, Texas	.77	.90
Baltimore, Maryland	.91	.89
Birmingham, Alabama	.79	.85
Bismark, North Dakota	.79	.94
Boise, Idaho	.83	.93
(Base) Boston, Massachusetts	1.00	1.00
Bridgeport, Connecticut	.98	1.01
Buffalo, New York	1.08	1.01
Camden, New Jersey	1.07	.97
Charleston, West Virginia	.94	1.00
Charlotte, North Carolina	.66	.93
Chattanooga, Tennessee	.81	.94
Cheyenne, Wyoming	.83	.94
Chicago, Illinois	1.07	.90
Cincinnati, Ohio	1.03	1.18
Cleveland, Ohio	1.10	.88
Columbus, Ohio	1.04	.95
Dallas, Texas	.75	1.06
Denver, Colorado	.86	.89
DesMoines, Iowa	.87	.97
Detroit, Michigan	1.21	.96
El Paso, Texas	.69	.90
Evansville, Illinois	.89	.94
Harrisburg, Pennsylvania	.92	.97
Hartford, Connecticut	1.00	.98
Honolulu, Hawaii	.92	1.02
Houston, Texas	.84	.91
Indianapolis, Indiana	.89	.94
Jackson, Mississippi	.72	.85
Jacksonville, Florida	.85	.95
Kansas City, Missouri	1.07	1.05
Lansing, Michigan	1.01	.93
Las Vegas, Nevada	1.04	1.06
Little Rock, Arkansas	.73	.86
Los Angeles, California	1.13	.97
Louisville, Kentucky	.95	.94

TABLE 3
(continued)

Location	Labor	Material
Madison, Wisconsin	.90	.95
Manchester, New Hampshire	.89	.97
Miami, Florida	1.02	.95
Milwaukee, Wisconsin	.99	.96
Minneapolis, Minnesota	.97	1.09
Mobile, Alabama	.84	.91
Nashville, Tennessee	.78	.90
Newark, New Jersey	1.12	.89
New Orleans, Louisiana	.83	.88
New York, New York	1.33	1.02
Norfolk, Virginia	.70	.94
Oklahoma City, Oklahoma	.83	.89
Omaha, Nebraska	.90	.97
Philadelphia, Pennsylvania	1.04	.78
Phoenix, Arizona	.98	.95
Pittsburgh, Pennsylvania	1.04	1.14
Portland, Maine	.71	.98
Portland, Oregon	.94	.95
Providence, Rhode Island	.97	.98
Richmond, Virginia	.71	.95
Rochester, New York	1.06	1.03
St. Louis, Missouri	1.04	.97
Salt Lake City, Utah	.88	.94
San Diego, California	1.02	.93
San Francisco, California	1.20	1.06
Savannah, Georgia	.77	.94
Seattle, Washington	.99	.74
Shreveport, Louisiana	.76	.86
Sioux Falls, South Dakota	.81	.94
Spokane, Washington	.91	.94
Springfield, Illinois	.89	.93
Springfield, Massachusetts	.98	.99
Syracuse, New York	1.00	1.00
Tampa, Florida	.88	.95
Topeka, Kansas	.86	.98
Trenton, New Jersey	1.09	1.02
Washington, D.C.	.99	.96
Wichita, Kansas	.92	.97
Wilmington, Delaware	.98	.97
Youngstown, Ohio	.99	.94
Montreal, Quebec	.73	.73
Toronto, Ontario	.94	1.09

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*1974 Dodge Guide for Estimating Public Works Construction Costs. Annual Edition No. 6, McGraw-Hill Information Systems Company, Princeton, New Jersey (1974)

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TABLE 4 SUMMARY OF TECHNIQUES AND COSTS

METHOD	SECTION OF REPORT	COSTS/ACRE		ADVANTAGES	DISADVANTAGES
		AVERAGE	RANGE		
1 RESTRICTION OF LAND USE (FENCING)	APPENDIX B, PART II	1100	350–3300	-INEXPENSIVE -LAND IS NOT DISTURBED OR DEGRADED	-LAND IS NOT PRODUCTIVE WHILE RESTRICTED -TECHNIQUES IS APPLICABLE FOR LOW ACTIVITY CONTAMINATION ONLY -REPLACEMENT OF FENCING IS NECESSARY AFTER 20 -30 YEARS
2 STABILIZATION A) CHEMICAL AND OR VEGETATIVE	TECHNIQUE 2 IN TABLE 1 AND 2	2100	500–3 000	-INEXPENSIVE -EASY TO APPLY -DOES NOT DISTURB THE LAND -LAND CAN BE PRODUCTIVE	-DOES NOT PREVENT PLANT UPTAKE OF PLUTONIUM -CHEMICALS MUST BE REAPPLIED EVERY 5 –10 YEARS UNLESS VEGETATIVE COVER IS ADEQUATE TO PREVENT RESUSPENSION OF PLUTONIUM -USED FOR RELATIVELY LOW ACTIVITY CONTAMINATION
B) PLOWING	TECHNIQUE 1 IN TABLE 1 AND 2	2 600	900 4,900	-INEXPENSIVE -PREVENTS RESUSPENSION OF PLUTONIUM PLOWING TO A DEPTH OF 3 FEET AIDS IN REDUCING PLANT UPTAKE OF PLUTONIUM -DILUTES THE PLUTONIUM ACTIVITY	-PLOWING MAKES FUTURE DECONTAMINATION EFFORTS VERY DIFFICULT -PLUTONIUM STILL REMAINS IN THE AREA -USED ON LOW OR MEDIUM LEVEL ACTIVITY CONTAMINATION
C) APPLICATION OF A SOIL COVER	TECHNIQUES 3A AND 3B IN TABLE 1 AND 2	4,800 (4 INCH COVER) 9,600 (12 INCH COVER)	2,400–7,500 5,400 - 14,000	-100% EFFECTIVE IN PREVENTING RESUSPENSION -LAND CAN BE PRODUCTIVE	-MODERATELY EXPENSIVE - THE MOST EXPENSIVE STABILIZATION TECHNIQUE -DOES NOT PREVENT PLANT UPTAKE (12 INCH SOIL COVER SHOULD BE MORE EFFICIENT IF PREVENTING PLANT UPTAKE THAN THE 4 INCH SOIL COVER
3 SOIL REMOVAL WITH ONSITE STABILIZATION A SCRAPE INTO WINDROWS	TECHNIQUES 4A OF TABLES 1 AND 2	3,600	1,600–6,300	-ABOUT 60% OF LAND IS ACTIVITY FREE (GENERALLY ABOUT 90% EFFICIENCY FOR REMOVAL OF PLUTONIUM) -ABOUT 60% OF LAND CAN BE PRODUCTIVE -RESUSPENSION AND PLANT UPTAKE OF Pu IS SIGNIFICANTLY REDUCED -FUTURE DECONTAMINATION EFFORTS WOULD NOT BE DIFFICULT SINCE ALMOST ALL OF THE ACTIVITY WOULD BE STABILIZED IN THE WINDROWS	-MODERATE COSTS -WINDROWS ARE STABILIZED BY CHEMICALS AND OR VEGETATION THUS, PROBLEMS ARE SIMILAR TO THOSE LISTED FOR 2, A ABOVE -ACTIVITY STILL REMAINS ONSITE
B) SCRAPE OR VACUUM INTO MOUNDS OR TRENCHES	TECHNIQUES 4B 4C AND 5 OF TABLES 1 AND 2	5,200	2200–10,200	-ABOUT 90% (OR BETTER) OF THE ACTIVITY IS REMOVED FROM THE LAND -ABOUT 90% OF THE LAND IS PRODUCTIVE -RESUSPENSION AND PLANT UPTAKE OF Pu IS SIGNIFICANTLY REDUCED -FUTURE DECONTAMINATION IF NECESSARY, WILL NOT BE DIFFICULT (HOWEVER IT WOULD BE A LITTLE MORE DIFFICULT THAN THE PREVIOUS CASE WHERE THE SOIL IS SCRAPED INTO WINDROWS)	-MODERATE COSTS -MOUNDS MAY REQUIRE FURTHER STABILIZATION OR DECONTAMINATION IN THE FUTURE -ACTIVITY STILL REMAINS ONSITE -LEACHING Pu INTO THE SOIL MAY BE A CONCERN WITH TRENCH DISPOSAL
4 SOIL REMOVAL WITH OFFSITE DISPOSAL	TECHNIQUES 6A AND 6B OF TABLES 1 AND 2	125,000 (WASTE BURIAL GROUND, NON RETRIEVABLE) 165,000 (WASTE BURIAL GROUND, RETRIEVABLE) 515,000 (FEDERAL REPOSITORY)	110,000-190,000 150,000–235,000 400,000 600,000	COMPLETE REMOVAL OF CONTAMINATED SOIL FROM THE AREA -PERPETUAL CARE OF THE CONTAMINATED SOIL AT A LICENSED BURIAL GROUND (IN RETRIEVABLE FORM THE SOIL MAY BE REPACKAGED, IF NECESSARY, AND MAINTAINED IN HIGH INTEGRITY CONTAINERS) NOTE SOIL WILL POSSIBLY NEVER BE DEPOSITED AT A FEDERAL REPOSITORY. CURRENT REQUIREMENTS ARE THAT THE ACTIVITY CONCENTRATION BE AT LEAST 10 nCi/gram BEFORE THE Pu CONTAMINATED SOIL IS DEPOSITED AT THE FEDERAL REPOSITORY	-EXTREMELY EXPENSIVE -SOIL MUST BE HANDLED MORE OFTEN THAN IN THE OTHER TECHNIQUES THUS INCREASING THE PROBABILITY OF AN ACCIDENTAL SPILL

TABLE 5

Plutonium Cleanup Experience Costs

Project	Total Cost	Acres	$/Acre
AEC Niagara Falls Site[a]	$ 88,000 (1972)	8-10	$ 8,800
Palomares, Spain[b]	$50,000,000 (1966)	600	$83,300
Nuclear Reactor Development[a] Station at NTS	$ 100,000 (1965)	5	$20,000
Enewetak Islands[c]	--- (1976)		$25,500*
Rocky Flats[d]		0.46	

Enewetak Islands Project Estimated Costs for 1976[c]

Technique	Actual Costs	Costs Adjusted for Continental U.S.	Costs Including 32% Overhead
		$/C.Y.	$/Acre*
Debrushing	$1,700.00/acre	---	$ 800
Scraping (including Stabilization)	6.40/cubic yard	2.3	1,620
Replacing Soil	11.00/cubic yard	3.9	2,790
Disposal	80.00/cubic yard	28.6	20,280
Totals	$ 97.4 /cubic yard	34.8	$25,500

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*Costs in $/C.Y. were converted to $/Acre by assuming that 4 inches of soil were removed or replaced. A 4-inch layer of soil is equal to 538 cubic yards of soil per acre. Actual costs are estimated to be about $70,000 per acre. The Army Corps of Engineers indicated that these costs are about a factor of 2.8 higher than the costs for decontaminating similar land in the United States.

[a] From reference 4 of this report. Note: Radioactivity deposited on the land was not Pu.

[b] Kathren, Ronald L. Towards Interim Acceptable Surface Contamination Levels for Environmental PuO. BNWL-SA-1510 (1968)

[c] Reference 31 of this report.

[d] Summary Report of Soil Removal - Preliminary Excavations, C.E. Wickland, Report presented at the Conference for Decontamination and Decommissioning (D and D) of ERDA Facilities held in Idaho Falls, Idaho, August 19-21, 1975.

TABLE 6. USES FOR STABILIZERS AND APPROXIMATE COSTS TO APPLY/ACRE

Temporary (prior to any decon)	Short-term (prior to soil removal)	Short-term (on bare soil as a part of restoration)	Long-term 5-10 years	Soil Cover
Water - $220 ($100-$340)				
	Asphaltic Emulsion - $1100			
	Geo-tech - $ 520		Geo-tech $520 (with reapplication every 5 years)	
	Dust control oil -$360-$730			
	Petroset $1200			
	Poly vinyl Acetate -$200-$700			
		Norlig - $240		
		DCA-70 $440		
			Rapid Cure Road Oil $660 (reapply after 5 years)	
				4" Loam Range $1500 - $3300 -Average $2400
				4" Topsoil Range $3300 - $5700 -Average $4500
Range $100-$340/acre Average $220/acre	Range $200-$1200/acre Average $700	Range $200-$1200 Average $570	Range $520-$660 Average $600	Loam Topsoil Range $1700-4500 +$3500-6900 Average $3100 $5200

TABLE 7

Value of Crops (per Acre)* in 1973

Crop	No. Units/Acre	Unit	$/Unit	$/Acre
Corn	91.4	bushels	2.37	217
Wheat	31.8	bushels	3.82	121
Soybeans	27.8	bushels	5.65	157
Oats (grain)	47.0	bushels	1.09	51
Tobacco	1983.0	pounds	0.90	1785
Hay	2.2	tons	40.60	89
All Crops (total U.S. Average)				170

Major crops - corn, soybeans, hay, wheat

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*U.S. Department of Commerce. Statistical Abstract of the United States 1974 National Data Book and Guide to Sources. Bureau of Census, Social and Economic Statistics Administration, U.S. Department of Commerce.

TABLE 8

Value of Forests (per Acre) in 1974*

Region	States	$/per 1000 Board Feet
1	Montana, N. Idaho	46.16
2	Colo., Wyoming, S. Dakota	12.65
3	Ariz., New Mexico	62.85
4	Utah, S. Idaho, Nevada	72.78
5	California	83.36
6	Oregon, Washington	124.35
8	S.E. (Va. to Texas)	50.76
9	Lake States and N.E.	17.21
10	Alaska	15.26
	Average for the U.S.	88.14

# board feet per acre --

National average	30,000
Region 6 average	60,000
Maximum	150,000

$ per acre --

National average	$ 2,644
Region 6	$ 7,461
Maximum (at $88.14/1000 board feet)	$13,200

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*U.S. Department of Agriculture. Timber Sold on National Forest, Fourth Quarter and Fiscal 1974. Circular Memorandum, July 22, 1975.

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